Fermi surface reconstruction and multiple quantum phase transitions in the antiferromagnet CeRhIn₅

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Conventional, thermally driven continuous phase transitions are described by universal critical behavior that is independent of the specific microscopic details of a material. However, many current studies focus on materials that exhibit quantum-driven continuous phase transitions (quantum critical points, or QCPs) at absolute zero temperature. The classification of such QCPs and the question of whether they show universal behavior remain open issues. Here we report measurements of heat capacity and de Haas-van Alphen (dHvA) oscillations at low temperatures across a field-induced antiferromagnetic QCP ($B_{c0} \approx 50$ T) in the heavy-fermion metal CeRhIn₅. A sharp, magnetic-field-induced change in Fermi surface is detected both in the dHvA effect and Hall resistivity at $B_0^* \approx 30$ T, well inside the antiferromagnetic phase. Comparisons with band-structure calculations and properties of isostructural CeCoIn₅ suggest that the Fermi-surface change at B_0^* is associated with a localized-to-itinerant transition of the Ce-4f electrons in CeRhIn₅. Taken in conjunction with pressure experiments, our results demonstrate that at least two distinct classes of QCP are observable in CeRhIn₅, a significant step toward the derivation of a universal phase diagram for QCPs.

heavy fermion | quantum phase transitions | superconductivity | Fermi surface reconstruction | localized-itinerant transition

Intermetallic heavy-fermion metals, the ground states of which can be tuned readily by control parameters other than temperature, such as magnetic field, pressure, or chemical composition, form attractive systems for the study of quantum critical points (QCPs) (1, 2). Despite examples of QCPs in many heavy-fermion materials, their theoretical classification, analogous to that for thermally driven phase transitions, is yet to be achieved (2, 3). An extension of the theory of thermally driven critical points to the zero-temperature limit predicts that only fluctuations of an order parameter [for example, the sublattice magnetization of a spin-density wave (SDW)] are singular in space and time at a critical value of the control parameter (4–6). The Fermi surface evolves smoothly across the QCP. Although there is some experimental support for this model of a QCP [e.g., in CeCu₂Si₂ (7)], the quantum critical response of some other heavy-fermion compounds is clearly inconsistent with its predictions (8–10). Going beyond this conventional Landau framework, a qualitatively different model predicts a sharp reconstruction of the Fermi surface while crossing the QCP owing to the essential involvement of the electronic degrees of freedom (11–13). Such an unconventional QCP has been proposed to involve the critical destruction of the Kondo effect, in addition to fluctuations of the order parameter (3). Behavior that supports this type of QCP has been found in various heavy-fermion systems, including UCu_{5-x}Pd_x (8), CeCu_{6-x}Au_x (9), and YbRh₂Si₂ (10). In each material, however, direct evidence for a change in Fermi surface is lacking. Besides the needs to verify the basic predictions of this model of QCPs, a further test of its validity is the more restrictive observation of a change in the Fermi surface as a function of multiple tuning parameters.

CeRhIn₅, a heavy-fermion antiferromagnet with a Néel temperature $T_N \approx 3.8$ K at ambient pressure (14), is very suitable for this purpose. As shown in the schematic phase diagram at zero temperature (Fig. 1A), application of pressure suppresses the antiferromagnetic (AF) order and induces superconductivity (15, 16). In the presence of a modest magnetic field, sufficient to suppress superconductivity, an AF QCP is exposed through pressure tuning (15, 16). A distinct change of the Fermi surface is observed across this pressure-induced QCP via dHvA oscillations (Fig. 1A, blue arrow) that find a large Fermi surface, like that of CeCoIn₅, above the QCP (17). In addition, the AF order of CeRhIn₅ is robust against magnetic field at ambient pressure (18). Thus, CeRhIn₅ permits measurements of magnetic quantum oscillations across its field-tuned QCP at ambient pressure, providing a rare system in which the nature of QCPs can be probed using more than one tuning parameter.

Here, we report our study of the phase transitions in CeRhIn₅ at ambient pressure using isothermal measurements of the *ac* heat capacity and dHvA oscillations in pulsed magnetic fields of up to 72 T (Fig. 1B), plus Hall resistivity and dHvA oscillations in a *dc* field of up to 45 T (SI Appendix). These experiments allow explicit mapping of the magnetic field-temperature phase diagram and the investigation of the evolution of the Fermi surface as a function of magnetic field.

Significance

Conventional, thermally driven continuous phase transitions are described by universal critical behavior that is independent of microscopic details of a specific material. An analogous description is lacking for phase transitions that are driven at absolute zero temperature by a nonthermal control parameter. Classification of quantum-driven phase transitions is a fundamental but open problem that arises in diverse contexts and multiple classes of materials. Here we report the first observation, to our knowledge, of a sharp Fermi surface reconstruction while applying a strong magnetic field to suppress an antiferromagnetic transition to zero temperature. These experiments demonstrate that direct measurements of the Fermi surface can distinguish theoretically proposed models of quantum criticality and point to a universal description of quantum phase transitions.

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200–1,000 T for fields of $10 \text{ T} < B < 30 \text{ T}$. They coincide well with our preceding data obtained in the 45-T hybrid magnet as well as with previous measurements (19) and are characteristic of a “small” Fermi surface that does not include the $4f$ electrons of Ce. Several higher harmonics of the most pronounced dHvA frequencies ($f_{a4} \approx 790 \text{ T}$) are observed. Four new dHvA branches (f_{a5} – f_{a8}) with much larger frequencies ($7 \text{ kT} < f < 15 \text{ kT}$) occur at $B > B^*$; the frequency of f_{a7} is close to that obtained in a dc field. These new dHvA frequencies persist up to $T = 0.8 \text{ K}$ but become difficult to resolve around $T = 1 \text{ K}$. It is noted that, owing to their short duration, pulsed experiments may give lower signal-to-noise ratios than equivalent dc field measurements. Hence, lower temperatures are needed to resolve all the dHvA oscillations.

Summary and Discussion

Fig. 5A presents the temperature–magnetic field phase diagram of CeRhIn_5 for $B//a$ and $B//c$ derived from our experimental data. On applying a magnetic field the AF transition temperature T_N is suppressed continuously to the lowest accessible temperature ($T \approx 0.4 \text{ K}$ for $B//a$), providing strong evidence for an AF QCP around $B_{c0} \approx 50 \text{ T}$. The field dependence of T_N , that is, $T_N \sim (B_{c0} - B)^{2/3}$, suggests an SDW-type QCP. Inside the AF phase the electronic system undergoes a sharp reconstruction of Fermi surface at $B^*(T)$, changing from a local-moment antiferromagnetic order (AF_S) to an SDW of the large Fermi surface (AF_L). Further experiments are needed to determine whether the B^* line eventually fades away in the paramagnetic state or terminates at a critical end point.

High-frequency dHvA oscillations potentially could emerge with increasing magnetic field owing either to the so-called Dingle damping factor or to magnetic breakdown (20). However, it is clear from Fig. 3 and Fig. 4A that the higher frequencies appear suddenly at B^* , rather than growing smoothly. If the Dingle factor were responsible for their growth, each oscillation should gradually become visible over differing ranges of field. The scenario of magnetic breakdown at B^* also can be excluded because the new dHvA frequencies appear at indistinguishable fields for $B//a$ and $B//c$; in an anisotropic band structure, such as that of CeRhIn_5 , any breakdown fields also would be anisotropic. Thus, all these suggest that the sudden emergence of new dHvA frequencies at B^* corresponds to a field-induced reconstruction of Fermi surface in CeRhIn_5 .

Modifications of the magnetic structure in a magnetic field (e.g., a metamagnetic transition) may induce a reconstruction of Fermi surface. With the current resolution of our experiments we cannot detect any thermodynamic phase transition at $B = B^*$ either in magnetic susceptibility or heat capacity (cf. Fig. 1B). Furthermore, the critical field of a metamagnetic transition usually varies with the field orientation as shown for the transition at B_M . Our observations of an isotropic value of B^* for $B//a$ and $B//c$ disfavor such a scenario. Therefore, it is unlikely that the field-induced Fermi surface reconstruction in CeRhIn_5 is caused by a change in magnetic structure. Nevertheless, it is desired to perform experiments (e.g., high-field NMR measurements) to directly examine it.

The delocalization of $4f$ electrons, resulting from Kondo screening, expands the Fermi volume and may give rise to the emergence of larger dHvA frequencies above B^* in CeRhIn_5 . This is supported by the following facts. First is evolution of dHvA frequencies: For $B < B^*$, previous dHvA studies have shown that the Ce- $4f$ electrons are localized in CeRhIn_5 and do not contribute to the Fermi sea (18, 21). Our results are compatible with this conclusion. Above $B^*(T)$ new dHvA frequencies are observed for $B//a$ and $B//c$ at low temperatures. They are comparable to both the ones measured in CeCoIn_5 (ref. 22 and SI Appendix, Fig. S6) and calculations for CeRhIn_5 under the assumption of itinerant $4f$ electrons (cf. SI Appendix, Fig. S5). Second is the change of the Hall coefficient: According to ref. 18,

Fig. 5. Experimental temperature–field phase diagram and its relation to a multiparameter theoretical phase diagram. (A) Temperature–magnetic field phase diagram of CeRhIn_5 at ambient pressure. Squares and circles represent the characteristic temperatures for $B//a$ and $B//c$, respectively. The Néel temperature, $T_N(B)$, was determined from the heat capacity measured in a PPMS with a dc field of 16 T (red) and in a pulsed field (yellow) as well as the dHvA oscillations (light blue, $B//a$). The dashed line displays the field dependence $T_N \sim (B_{c0} - B)^{2/3}$, expected for a 3D-SDW QCP (6). The field $B^*(T)$ is determined from a jump of the Hall resistivity (orange circles, $B//c$) and the onset of new dHvA frequencies in the Fourier transform spectra (white square for $B//a$; white circles for $B//c$). The error bars were determined from the width of the Hall resistivity jumps and the field interval $\Delta B = 5 \text{ T}$ used in the Fourier transform analysis. The phases AF_S , AF_L , and P_L represent AF or paramagnetic (P) states with large (L) or small (S) Fermi surfaces. (B) A schematic magnetic field–pressure phase diagram of CeRhIn_5 at $T = 0$. A pressure-induced AF QCP exists at low field and is accompanied by a sharp change of the Fermi surface, indicating a destruction of the Kondo effect at zero temperature (17). However, as shown in this work, a sufficiently strong magnetic field at ambient pressure also continuously suppresses the AF phase at B_{c0} . In this case, the Kondo destruction takes place inside the AF state, and the AF QCP is likely to be of an SDW type. Further measurements are to be performed to determine experimentally the phase diagram in the regime of finite pressures and high magnetic fields.

the Fermi surface of LaRhIn_5 , which has no $4f$ electrons, is dominated by electron pockets; the Fermi surface of CeRhIn_5 will be similar if the $4f$ electrons are localized, accounting for the negative Hall coefficient at fields below B^* . However, in CeCoIn_5 the $4f$ electrons are itinerant, and the Fermi surface is more 3D; the volumes of electron and hole pockets are nearly compensated (18). With delocalized $4f$ electrons the Fermi surface of CeRhIn_5 would be of an analogous form, explaining the more complicated variation of the Hall coefficient above B^* . We note that a change in magnetic structure (e.g., a metamagnetic transition as discussed above) may lead to a jump in the Hall

resistivity. However, the Hall number usually is unchanged across a metamagnetic transition, as observed at B_M for CeRhIn_5 (Fig. 3A). This adds further evidence against a change of magnetic structure at B^* . Third is enhancement of the heat-capacity coefficient: In Fig. 2B, *Inset*, we have shown that the heat-capacity coefficient, $C_p(B)/T$, at low temperatures is substantially enhanced upon applying a magnetic field to suppress the AF order, suggesting the development of a Kondo-derived heavy electron state in a magnetic field. The persistence of the large Fermi surface up to at least $B = 70$ T at low temperatures can be understood by a simple consideration of the magnetic field and temperature scales. The temperature for the onset of Kondo screening in CeRhIn_5 is about 10 K (14), more than twice the Néel temperature. Given that the critical magnetic field B_{c0} is about 50 T, the single-ion Kondo field is expected to be substantially larger than the presently accessible fields.

The above analyses suggest that, upon increasing the magnetic field at ambient pressure and zero temperature, (i) an abrupt change of Fermi surface associated with a localized-itinerant transition of the $4f$ electrons occurs at $B^* \approx 30$ T (i.e., inside the AF state of CeRhIn_5), whereas (ii) the AF QCP at $B_{c0} \approx 50$ T is presumably of the SDW type. This is in contrast to what happens as a function of pressure at relatively low magnetic fields, where the dHvA oscillations indicate a $4f$ -localized/itinerant transition at the AF QCP (17). Our results are therefore twofold. First, there are at least two different types of quantum phase transitions in CeRhIn_5 , one induced by tuning a magnetic field and the other accessed with varying pressure. Moreover, under the tuning of these two parameters, a jump of Fermi surface occurs inside the AF state and at the AF QCP, respectively.

As a potential scenario, in Fig. 5B we present a schematic magnetic field-pressure phase diagram for CeRhIn_5 at zero temperature. Such a phase diagram can be qualitatively interpreted in terms of a global phase diagram of quantum-critical heavy-fermion metals (23, 24). This model delineates the evolution of the zero-temperature AF transition and the Kondo destruction in a multiparameter phase space. While tuning the compound by a magnetic field at zero pressure, the Kondo resonances are switched on at $B_0^* [=B^*(T \rightarrow 0)]$ before the AF order is suppressed at B_{c0} , and the Fermi surface evolves smoothly across the latter transition. This is in contrast to the transition caused by pressure at relatively low field, at which the Kondo destruction and magnetic transition take place simultaneously, leading to a jump of the Fermi surface at the zero-temperature continuous AF phase transition (17). We note that the dynamical Kondo effect makes the mass heavy even when the Fermi surface is small (3, 23). According to this model, the AF QCP associated with the pressure-induced superconducting dome is then likely to be of the unconventional type. This indicates that heavy-fermion superconductivity not only arises in the vicinity of SDW-type QCPs (7, 25) but can also be driven by electronic fluctuations arising from such an unconventional QCP at which the Kondo effect is destroyed.

Our experimental results and understandings have implications beyond CeRhIn_5 . They can be connected to the disparate results in a variety of heavy-fermion materials. Considerable

evidence already exists that pure YbRh_2Si_2 under magnetic-field tuning (10) and $\text{CeCu}_{6-x}\text{Au}_x$ under the variation of doping (9) feature Kondo destruction at their respective AF QCP. However, there also is evidence for Kondo destruction inside the AF region of Co-doped YbRh_2Si_2 (26) as well as in magnetic-field-tuned $\text{CeCu}_{6-x}\text{Au}_x$ (27) and $\text{Ce}_3\text{Pd}_{20}\text{Si}_6$ (28). In CeIn_3 , dHvA frequencies and the corresponding cyclotron masses of heavy-hole pockets undergo a sharp increase near 40 T, which is below the critical field needed to suppress AF order to a QCP (29). This suggests the possibility of a similar localization/itinerant transition inside its AF phase, even though dHvA oscillations associated with the large Fermi surface have not yet been observed at high fields. In a related vein, it has been suggested that Kondo destruction occurs outside the AF region in materials such as Ir-doped YbRh_2Si_2 (26) and $\text{Yb}_2\text{Pt}_2\text{Pb}$ (30). Our Fermi-surface characterization of multiple quantum phase transitions in a single compound not only provides new understanding about CeRhIn_5 but also strengthens the case that the quantum phase transitions of the other electron-correlated materials may be placed on the proposed global phase diagram.

Methods

A detailed description of the methods and materials is given in *SI Appendix*. Single crystals of CeRhIn_5 were grown by a flux method. Room-temperature powder X-ray diffraction measurements revealed that all of the crystals are single-phase and crystallize in the tetragonal HoCoGa_5 structure. The orientation of the crystal was determined by X-ray Laue diffraction (*SI Appendix*, section 1). Heat capacity and dHvA oscillations were measured in pulsed field magnets at the Los Alamos National Laboratory Pulsed Field Facility (*SI Appendix*, section 2). A typical sample size for this study is about $1 \text{ mm} \times 0.5 \text{ mm} \times 0.1 \text{ mm}$. An ac calorimeter was used for heat capacity measurements at fields up to $B = 53$ T and temperatures down to $T = 0.9$ K. Subtractions of the field-independent addenda contributions from the total heat capacity allow us to determine the absolute values of heat capacity for the sample. Magnetic susceptibility was measured by an induction method up to $B = 72$ T and at temperatures down to $T = 0.4$ K, obtained by pumping on liquid ^3He in a ^3He bath cryostat. The transverse Hall resistivity $\rho_{xy}(B)$ as well as the dHvA oscillations based on a torque method were measured in a ^3He cryostat and in fields to 45 T generated by the hybrid magnet at the National High Magnetic Field Laboratory (*SI Appendix*, sections 3 and 4). Band structure calculations were performed using the full-potential linearized augmented plane wave method as implemented in the WIEN2k code (*SI Appendix*, section 5).

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