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Anisotropic magneto-transport properties of the heavy fermion superconductor CeRh_2As_2

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We report anisotropic resistivity measurements of the heavy fermion superconductor CeRh_2As_2 in magnetic fields up to 16 T and temperatures down to 0.35 K. The measured CeRh_2As_2 resistivity shows a signature corresponding to the suggested quadrupole density wave order state at $T_0 \sim 0.5$ K for both measured directions. For a magnetic field applied along the tetragonal a axis, T_0 is enhanced with magnetic field reaching ~ 1.75 K at 16 T. Further, a magnetic field-induced transition occurs at $\mu_0 H_m \sim 8.1$ T corresponding to a change to a new broken symmetry state. For a magnetic field applied along the c axis, T_0 is suppressed below our base temperature ~ 0.35 K by $\mu_0 H \sim 4.5$ T, a field close to the previously reported field-induced transition within the superconducting state suggested to be from an even-parity to an odd-parity state. Our results indicate that the multiple superconducting phases in CeRh_2As_2 are intimately tied to the suppression of the proposed quadrupole-density-wave phase at T_0 .

A. Introduction

Systems exhibiting multiple superconducting phases are a rarity in nature. Only a handful of such compounds are known to exist, prominent among them are UPt_3 [1], thorium-doped UBe_{13} [2], $\text{PrOs}_4\text{Sb}_{12}$ [3–5] and UTe_2 [6–8]. The multiple superconducting phases in these systems are proposed to be useful in the pursuit of topological quantum computation. The recently discovered heavy fermion superconductor CeRh_2As_2 is a recent addition to this exotic class of systems [9].

CeRh_2As_2 crystallizes in the CaBe_2Ge_2 -type centrosymmetric tetragonal crystal structure (space group $P4/nmm$) [10] as shown in Fig. 1(a). It becomes superconducting (SC) below $T_c = 0.26$ K. The upper critical magnetic fields along both the principal axes (i.e. $\mu_0 H \parallel a \approx 2$ T and $\mu_0 H \parallel c \approx 14$ T) well exceed the Pauli paramagnetic limit $\mu_0 H_{PPL} = 1.86(\text{T/K})T_c \sim 0.5$ T. For a magnetic field applied along the c -axis, a field-induced transition occurs within the superconducting state at $\mu_0 H^* \sim 4$ T, which has been suggested to correspond to a change of the superconducting state from an even-parity (SC1) to an odd-parity state (SC2). In contrast, for a field applied in the ab -plane only one superconducting phase exists [9, 11]. Interestingly, nuclear quadrupole resonance measurements suggest an antiferromagnetic order also exists within the superconducting state [12].

Another intriguing second-order phase transition at $T_0 \sim 0.4$ K was identified precursing the superconducting state and was suggested to be a non-magnetic quadrupole-density-wave (QDW) phase (I) [13]. A feature corresponding to T_0 was observed in specific heat, thermal expansion and resistivity [9, 13]. For a magnetic field applied in the ab -plane, a field-induced transition to a new phase was observed at $\mu_0 H_m \sim 9$ T

in magnetostriction, resistivity, magnetization and magnetic torque [13].

The origin of the multiple superconducting phases due to the field-induced transition at $\mu_0 H^* \sim 4$ T in CeRh_2As_2 remains elusive. Currently, the unique crystal structure of CeRh_2As_2 is believed to lie at the heart of its unique superconducting properties. In CeRh_2As_2 , although the global inversion symmetry is preserved, the local inversion symmetry is lacking as Ce atoms have different Rh and As environments above and below it. This enables a staggered Rashba spin-orbit coupling and is suggested to be responsible for the field-induced transition within the superconducting state [9]. Thus, its nonsymmorphic crystal structure is suggested to play a key role in the field-induced transition [14, 15]. Khim et al [9], however, did not exclude the possibility of normal state properties affecting the superconducting properties in CeRh_2As_2 . They pointed out that the feature in specific heat at T_0 also seemed to be suppressed by a field $\mu_0 H \sim 4$ T. The specific heat feature, however, is subtle and therefore requires more careful studies.

Nonetheless, the rich phase diagram of CeRh_2As_2 raises the question of the relationship between the multiple phases, especially the superconducting state and the suggested QDW phase below T_0 . In this regard, the unique behaviour of the superconducting phase in applied magnetic fields along the two principal crystallographic directions i.e. a - and c -axes warrants a similar anisotropic investigation of the suggested QDW phase below T_0 . Furthermore, in the previous transport study [13], the current was applied in the basal plane of the tetragonal structure based on which it was suggested that the propagation vector of the order parameter of the phase below T_0 has a component within the basal plane. To the best of our knowledge, transport studies with current applied perpendicular to the basal plane i.e., along the c axis are still lacking. Therefore, an important piece of information regarding the order parameter of the T_0 phase remains missing. To remedy these shortcom-

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ings, we performed anisotropic magneto-transport measurement on microstructured devices made out of a single crystal of CeRh_2As_2 .

In this paper, we present anisotropic magneto-transport measurement in CeRh_2As_2 . Our main findings are : 1. CeRh_2As_2 is weakly anisotropic with the in-plane resistivity being roughly three-fourths the inter-layer resistivity. 2. A signature corresponding to the suggested QDW phase at T_0 is also observed in the out-of-plane resistivity. 3. For a magnetic field applied along the c -axis, T_0 is suppressed below our base temperature ~ 0.35 K by a magnetic field $\mu_0 H \sim 4.5$ T, a field, possibly coincidentally, similar to the previously reported field-induced even-odd parity transition within the superconducting state.

B. Experimental details

Single crystals of CeRh_2As_2 were grown in Bi flux starting from a mixture of pure elements Ce, Rh, As, and Bi with a molar ratio of 1 : 2 : 2 : 30. Starting materials were sealed in an evacuated fused silica tube, which was heated to 1150° over 30 h, followed by a dwell at 1150° for 24 h, then gradually cooled to 700° at a rate of $2.5^\circ/\text{h}$. The crystallographic structure of CeRh_2As_2 was verified at room temperature by a Bruker D8 Venture single-crystal X-ray diffractometer equipped with Mo radiation. X-ray diffraction analysis shows that CeRh_2As_2 crystallizes in the tetragonal space group $P4/nmm$ (No. 129) with lattice parameters $a = b = 4.28$ Å and $c = 9.85$ Å, in agreement with a previous report [9]. The transport measurements were performed using a Quantum Design Physical Property Measurement System (QDPPMS) equipped with a ^3He option reaching a base temperature of ~ 350 mK. For zero-field measurements an adiabatic demagnetization refrigerator extended the base temperature down to ~ 100 mK. The four-wire resistance was measured using an AC resistance bridge (Lakeshore model 372). The anisotropic-transport measurements were performed on a microstructured device of CeRh_2As_2 fabricated using focused ion-beam (FIB) milling from a single crystal as shown in Fig. 1 (b). After Laue-orienting the single crystal, it was polished along the ac -plane down to a thickness of $35\text{ }\mu\text{m}$. Out of the oriented single crystal, a six-terminal L-shaped microstructured device was fabricated to measure the anisotropic electrical transport properties along the two bar-shaped sections aligned with crystallographic a (D_a) and c (D_c) axes as shown in Fig. 1(b). The dimensions ($l \times w \times h$) of the two bar-shaped sections with current along a and c axes, namely D_a and D_c , are $180\text{ }\mu\text{m} \times 35\text{ }\mu\text{m} \times 35\text{ }\mu\text{m}$ and $120\text{ }\mu\text{m} \times 35\text{ }\mu\text{m} \times 35\text{ }\mu\text{m}$, respectively. The current and voltage leads for the device are labelled in Fig. 1 (b).

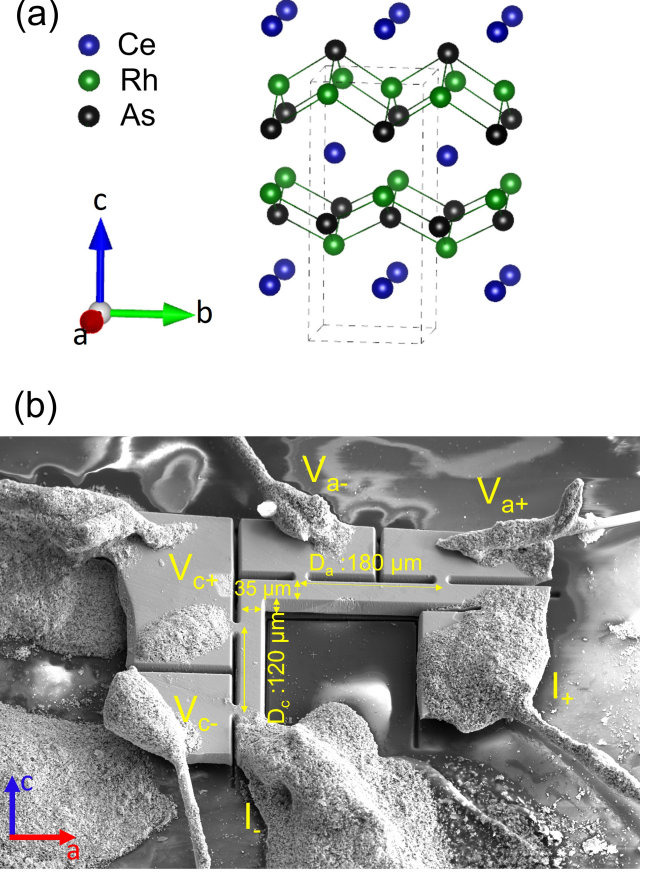


FIG. 1. (a) CaBe_2Ge_2 -type tetragonal crystal structure of CeRh_2As_2 with the elements Ce, Rh and As labelled in blue, green and black, respectively. The tetragonal unit cell is depicted by the dashed box. (b) FIB-fabricated microstructured device on a CeRh_2As_2 single crystal. D_a and D_c represent the two bar-shaped sections of the device, aligned along the crystallographic a - and the c -axes, respectively. Current and voltage leads as well as the device dimensions are labelled.

C. Results

Fig. 2(a) shows the anisotropic resistivities $\rho_a(T)$ and $\rho_c(T)$ of CeRh_2As_2 for a current $I = 10\text{ }\mu\text{A}$ applied in the microstructured device along the two bar-shaped sections D_a and D_c i.e., along the a - and the c -axes of the tetragonal system, respectively. The superconducting transition, marked by a drop to the zero resistivity state, occurs at $T_c \sim 0.26$ K in agreement with that reported through bulk thermodynamic probes [9, 11, 13]. The superconducting transition is sharper in $\rho_c(T)$ compared to $\rho_a(T)$. CeRh_2As_2 shows the typical resistive behaviour of a heavy fermion system. The broad hump-like feature in both $\rho_a(T)$ and $\rho_c(T)$ around ~ 40 K corresponds to the development of a coherent Kondo lattice. As evident from Fig 2(a), the in-plane resistivity (ρ_a) is roughly three-fourths the inter-layer resistivity (ρ_c),

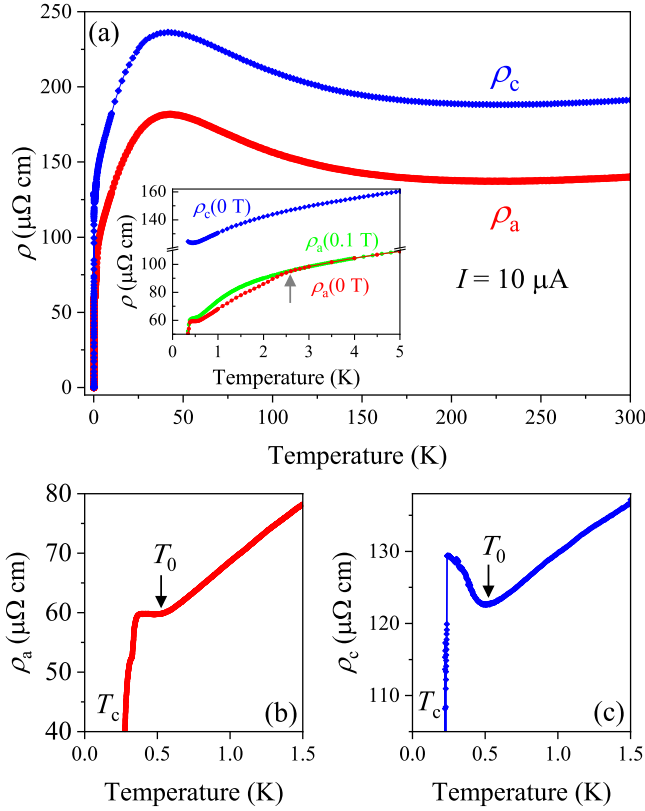


FIG. 2. (a) Resistivities ρ_a and ρ_c for a current $I = 10 \mu\text{A}$ applied to the microstructured sections D_a and D_c , respectively. Inset shows the resistivities $\rho_a(0 \text{ T})$, $\rho_a(0.1 \text{ T})$ and $\rho_c(0 \text{ T})$. Panel (b) and (c) shows a zoomed in view of the low-temperature resistivities ρ_a and ρ_c , respectively.

implying that the transport properties of CeRh_2As_2 are weakly anisotropic.

The inset in Fig. 2(a) shows resistivities $\rho_a(T)$ and $\rho_c(T)$ measured at zero-field as well as $\rho_a(T)$ measured at $\mu_0 H = 0.1 \text{ T}$. The grey arrow points to the resistance drop at $\sim 2.5 \text{ K}$ due to inclusion of a bismuth rich superconducting impurity as determined by energy-dispersive X-ray spectroscopy in the D_a bar. This resistance drop vanishes in a field as small as 0.1 T . The impurity can be seen as a light patch in SEM image of Fig. 1(b). No such impurity inclusion exists in the D_c bar.

Below the coherence temperature, $\rho_a(T)$ and $\rho_c(T)$ decrease rapidly down to $T_0 \sim 0.5 \text{ K}$. Below T_0 , $\rho_a(T)$ shows little temperature-dependence down to the superconducting transition. In contrast, $\rho_c(T)$ exhibits a strong upturn and continues to increase down to the superconducting transition [see Fig. 2(b) and (c)]. For $\rho_a(T)$, we define T_0 as the temperature where $\partial^2 \rho_a(T)/\partial T^2$ has a local extremum as marked by an arrow in Fig. 2(b). In $\rho_c(T)$, this corresponds to the local minima before the strong upturn and therefore we take the local minimum as T_0 for $\rho_c(T)$, as marked by arrow in Fig. 2(c).

Previously, a suggestion of a gap opening at the Fermi level was made based on the behaviour of $\rho_a(T)$ below

T_0 [13]. This signature of T_0 in $\rho_a(T)$, in conjunction with its behavior observed in other measurements, was suggested to be an indication of a QDW state [13]. Similarly, our observation of the strong increase in $\rho_c(T)$ below T_0 suggests the opening of a gap at the Fermi level. Furthermore, the presence of a strong upturn in $\rho_c(T)$ in comparison to $\rho_a(T)$ at T_0 suggests that the propagation vector of the order parameter of the phase below T_0 also has an out-of-plane component, in addition to an in-plane component reported previously [13].

Next, to determine the field-evolution of the phase below T_0 , we measured $\rho_a(T)$ and $\rho_c(T)$ at several constant magnetic fields up to 16 T applied along both the principal axes of the tetragonal system i.e. a - and c -axes as shown in Fig. 3. The open and solid black arrows point to the feature at T_0 at different fields. It is clearly evident from Fig. 3 that T_0 evolves differently for the two field orientations. For a same field orientation i.e., $H \parallel a$ or $H \parallel c$, T_0 evolves similarly in both $\rho_a(T)$ and $\rho_c(T)$. For $H \parallel a$, a clear distinction is evident in both $\rho_a(T)$ and $\rho_c(T)$ at fields above and below 8 T as marked by solid black ($\mu_0 H \leq 8 \text{ T}$) and open black ($\mu_0 H \geq 8 \text{ T}$) arrows in Fig. 3(a) and (b). The subtle plateau in $\rho_a(T)$ and the upturn in $\rho_c(T)$ vanish near 8 T and becomes a downturn consistent with a transition into a new phase above 8 T . Therefore, above 8 T , T_0 is obtained from the local minimum in $\partial^2 \rho_c(T)/\partial T^2$ and $\partial^2 \rho_c(T)/\partial T^2$. In contrast, for $H \parallel c$, T_0 is monotonously suppressed with field and there is no indication of a phase transition down to 0.35 K for $\mu_0 H \geq 4.5 \text{ T}$, as shown in Fig. 3(c) and (d). Measurements to lower temperatures are necessary to uncover the fate of T_0 in relation to superconductivity.

To further elucidate the phase diagram, we measured the anisotropic magnetoresistances i.e., $\rho_a(\mu_0 H)$ and $\rho_c(\mu_0 H)$ at several constant low-temperatures for fields applied along the a - and the c -axes of CeRh_2As_2 as shown in Fig. 4. For field applied along the a -axis, there is distinct field-induced transition occurring at $\mu_0 H_m \sim 8.1 \text{ T}$ for temperatures below $\sim 1.2 \text{ K}$ as shown in Fig. 4 (a) and (b). The transition is sharper in $\rho_a(\mu_0 H)$ relative to $\rho_c(\mu_0 H)$ possibly suggesting the states that contribute more to the in-plane electronic properties are more strongly affected by the phase transition. For fields applied in the ab -plane, a field-induced transition was previously observed at $\sim 9 \text{ T}$ and was suggested to correspond to a change from QDW (I) to another non-magnetic phase (II) [13]. In addition, there is another subtle feature in both $\rho_a(\mu_0 H)$ and $\rho_c(\mu_0 H)$ at $\mu_0 H_0 \sim 2.5 \text{ T}$ at 500 mK . Its temperature evolution is marked by arrows in Fig. 4(a) and (b), respectively. On the other hand, for field applied along the c axis, no such field-induced transitions are apparent in either $\rho_a(\mu_0 H)$ or $\rho_c(\mu_0 H)$. The sharp increase in $\rho_a(\mu_0 H)$ at very low fields for both $H \parallel a$ and $H \parallel c$ is extrinsic to CeRh_2As_2 and attributed to a superconducting impurity inclusion discussed previously.

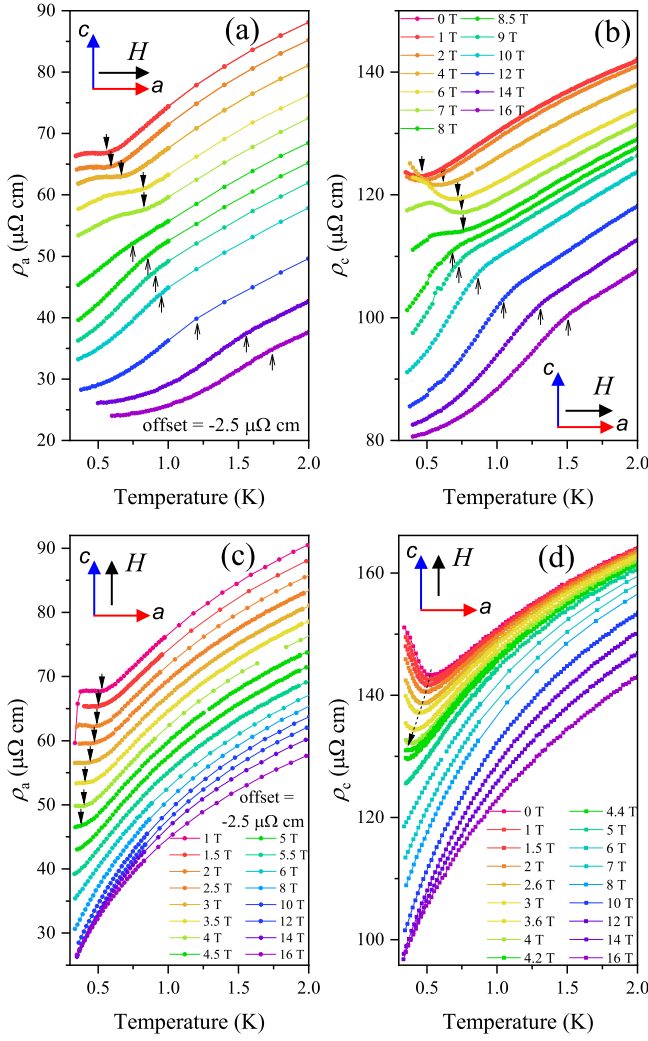


FIG. 3. (a) $\rho_a(T)$ and (b) $\rho_c(T)$ at different constant fields applied along the a -axis of CeRh₂As₂. Solid (open) black arrows point to the T_0 feature below (above) 8 T. Panels (c) and (d) shows the corresponding plots for field applied along the c -axis of CeRh₂As₂. Black arrows point to the feature at T_0 . In panel (a) and (c), the curves are shifted vertically for clarity. The legend for panel (a) is the same as panel (b).

D. Discussion

We plot a revised temperature-magnetic field ($T-H$) phase diagram for CeRh₂As₂ based on Fig. 3 and 4, along with data from ref [9] and [13], as shown in Fig. 5. Our phase diagram for $H \parallel a$ agrees well with ref [13]. The proposed QDW phase that exists below $T_0 \sim 0.5$ K at zero field extends to higher temperatures with increasing fields. At the highest field of our measurement i.e., 16 T, T_0 occurs at ~ 1.75 K. Furthermore, a clear field-induced transition occurs at $\mu_0 H_m \approx 8.1$ T corresponding to a change from the suggested QDW state to a new broken symmetry state below T_0 . A second phase transition exists at $\mu_0 H_0$ corresponding to the T_0 feature

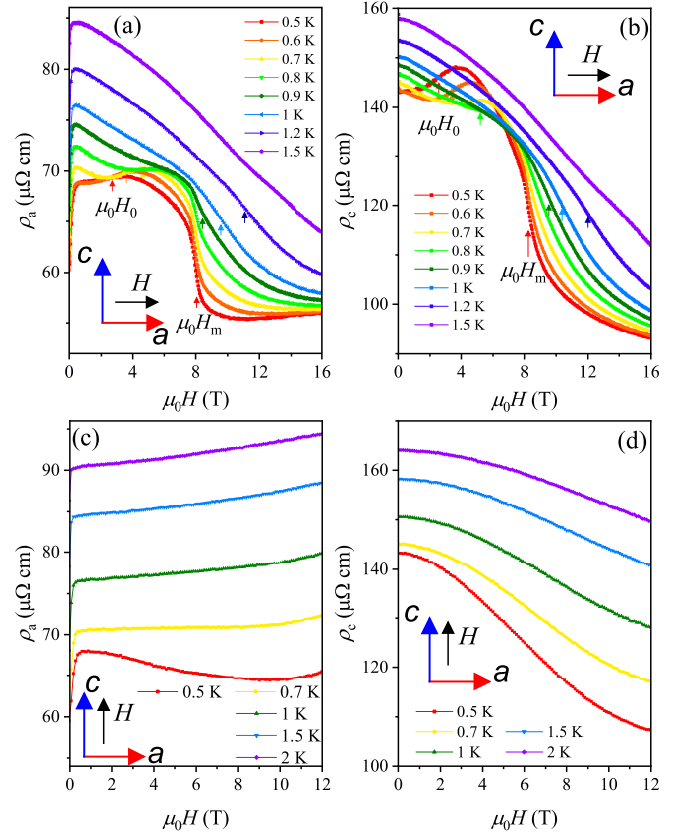


FIG. 4. (a) $\rho_a(\mu_0 H)$ and (b) $\rho_c(\mu_0 H)$ at several constant low-temperatures for a field applied along the a -axis of CeRh₂As₂. The field-induced transitions $\mu_0 H_m$ and $\mu_0 H_0$ are marked by arrows. (c) $\rho_a(\mu_0 H)$ and (d) $\rho_c(\mu_0 H)$ at several constant low-temperatures for a field applied along the c -axis of CeRh₂As₂.

in $\rho_a(\mu_0 H)$ and $\rho_c(\mu_0 H)$.

The most interesting result is obtained for $H \parallel c$. In both $\rho_a(T)$ and $\rho_c(T)$, the feature at T_0 is suppressed below our base temperature ~ 0.35 K by a field $\mu_0 H \sim 4.5$ T. This suppression field is close to the one for the field-induced transition from even-parity (SC1) to odd-parity (SC2) superconducting state shown by open circles in Fig. 5(b) based on Ref [9] which may be a coincidence.

Based on this observation, we envision two scenarios for how the proposed QDW phase might create multiple superconducting phases for which lower temperature measurements are required. These scenarios are depicted in Fig. 5(c). First, if the T_0 phase boundary indeed meets at the multicritical point [see Fig. 5(c)(i)], this would suggest that SC1 is a phase for which SC and the proposed QDW phase below T_0 coexist, while SC2 possesses only SC order. Such a scenario would also allow us to infer the order of the field-induced transition within the superconducting state. Since it is thermodynamically forbidden for three second-order phase transition lines to meet at a tri-critical point [16], the field-induced transition was thought to be first-order [9]. But T_0 phase boundary merging at the multi-critical point would re-

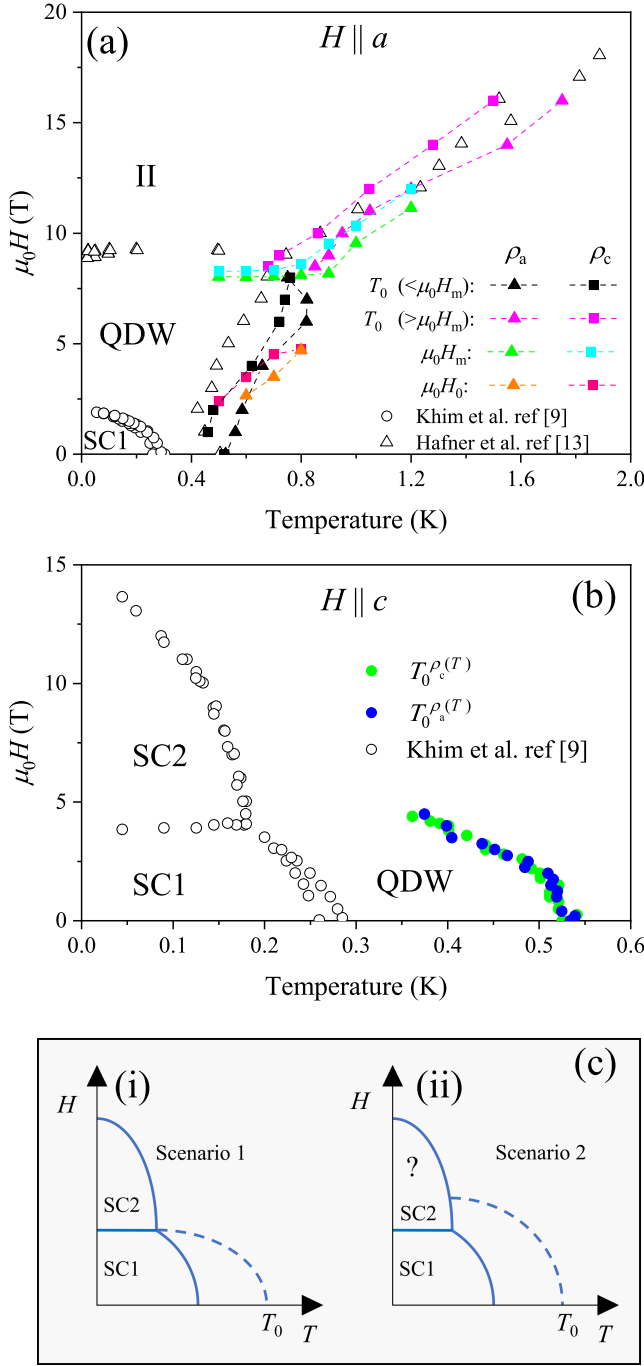


FIG. 5. Temperature-magnetic field ($T-H$) phase diagram for CeRh_2As_2 based on our magneto-transport measurements for field applied along (a) the a - and (b) the c -axes. In panel (a) magenta (solid black) symbols correspond to T_0 features in $\rho(T)$ at field above (below) $\mu_0 H_m$. Solid triangles (squares) correspond to the features in ρ_a (ρ_c). Green triangles (cyan squares) correspond to the feature at $\mu_0 H_m$ in $\rho_a(\mu_0 H)$ ($\rho_c(\mu_0 H)$). In panel (b) blue (green) solid circles correspond to the T_0 features in $\rho_a(T)$ ($\rho_c(T)$). The data corresponding to the open circles are taken from Ref [9] and the open triangles is taken from Ref [13]. (c) Two scenarios for T_0 phase boundary as discussed in the text.

move this thermodynamic constraint and allow the field-induced transition to be second-ordered. Alternatively, we can envision the T_0 phase boundary terminating at SC2 for a field above the multicritical point [see Fig. 5(c)(ii)]. This would suggest a scenario where the fluctuations associated with a field-induced QDW quantum critical point (QCP) mediate the SC2 phase similar to how SC is often found in the vicinity of an antiferromagnetic QCP [17, 18]. It is worth noting that one might expect evidence for a phase boundary within SC2. Otherwise, this scenario is thermodynamically forbidden [16] under the assumption that the proposed QDW transition is second-ordered in field. As stated above, lower temperature measurements are needed to distinguish between these two scenarios.

E. Summary

In summary, we performed anisotropic magneto-transport measurement in the heavy fermion superconductor CeRh_2As_2 . We find that the proposed quadrupole density wave phase at T_0 manifests in both $\rho_a(T)$ and $\rho_c(T)$. Furthermore, for a magnetic field applied along the a -axis, T_0 rises with increasing magnetic fields and a field-induced transition occurs at $\mu_0 H_m \approx 8.1$ T where the suggested quadrupole density wave phase changes to a new broken symmetry state. In comparison, for a magnetic field applied along the c -axis, the quadrupole density wave phase at T_0 is suppressed below our base temperature ~ 0.35 K by a field $\mu_0 H \sim 4.5$ T, close to the transition field between two different SC phases. Our results suggest that the quadrupole density wave phase plays a key role in the unique superconducting properties of CeRh_2As_2 and leads to multiphase superconductivity when a magnetic field is applied along the c -direction.

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