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New quantum phase in the heavy fermion superconductor $CeRhIn_5$

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Abstract

Pressure dependence on the specific heat of $CeRhIn_5$ has been studied for magnetic field perpendicular to the ab-plane. Similar to the field parallel to the ab-plane [T. Park et al., Nature 440 (2006) 65], magnetic field reveals hidden magnetism in the mixed superconducting state above 1.75 GPa where antiferromagnetism disappears at zero field. The specific heat anomaly at T_c is initially suppressed with increasing field, but becomes enhanced with further increasing field, indicating that the nature of the superconducting (SC) transition may have changed from second to first order. Together with the coexisting phase of the field-induced magnetism and superconductivity, the first-order like SC transition in $CeRhIn_5$ may provide insight to the inhomogeneous SC state (or FFLO state) observed in $CeCoIn_5$.

Keywords: Quantum phase transition; $CeRhIn_5$; Heavy fermion superconductor; Hidden magnetism

1. Introduction

$CeRhIn_5$ is a heavy-fermion antiferromagnet (AF) with incommensurate, helical magnetic structure at ambient pressure and evolves with applied pressure to become a heavy-fermion superconductor [2]. The relationship between antiferromagnetism and superconductivity and the relatively high transition temperatures of both broken symmetries make $CeRhIn_5$ an ideal candidate to explore these states over wider temperature and pressure scales. With increasing pressure, the Neel temperature increases from 3.7 K and passes through a maximum near 0.5 GPa, where bulk superconductivity starts to appear [3]. The superconducting transition temperature T_c increases with pressure and exceeds the T_N near 1.75 GPa ($=P_1$), where signatures for the AF transition are lost. Recently, NQR measurements showed that AF and SC states coexist below the critical pressure P_1 [4]. At 2.35 GPa ($=P_2$), de Hass van Alphen (dHvA) measurements at high magnetic fields when superconductivity is suppressed shows divergence of the effective mass and the Fermi surface volume expansion [5]. In the normal state, the temperature dependence of the electrical resistivity shows T-linear behavior [6] and electronic specific heat C/T diverges as $T \to 0$ K [1]. These novel features in the normal state above the superconducting dome, which is typically associated with a quantum critical point (QCP), seem inexplicable because the plausible QCP candidate P_1 is not only first-order like, but also is well below P_2.

Recent pressure study of the specific heat of $CeRhIn_5$ for $H \| ab$-plane revealed field-induced magnetism in the mixed superconducting state for $P_1 < P < P_2$ [1]. In the H–P phase diagram, the second-order quantum phase boundary between the pure superconducting state and the coexisting phase of superconductivity and magnetism evolves with pressure from $H = 0$ kOe at 1.75 GPa to H_{c2} (=88 kOe) at 2.35 GPa, therefore naturally explaining the non-Fermi liquid behavior in the normal state and the phase transition from localized to delocalized f-electron state at 2.35 GPa. In high-T_c cuprates, especially in the parenting compound $La_{2-x}Sr_xCuO_4$, neutron scattering measurements showed that long-range antiferromagnetic (AFM) order is completely suppressed near the hole doping concentration $x_1 \approx 0.12$, suggesting an AFM QCP inside the superconducting dome [7]. At higher doping concentration, magnetic field induces an AFM order in the

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mixed superconducting state. The magnetic quantum phase line evolves with hole doping from $H = 0$ kOe at x_1 to, possibly, H_{c2} at the optimal doping x_2 (=0.163), where signatures for a quantum critical point have been observed in the normal state [8]. The presence of magnetic quantum critical point veiled by superconducting dome, which seems an ubiquitous phenomenon in magnetically mediated superconductors, suggests that quantum fluctuations may provide the bonding to form Cooper pairs. Here, we present extensive pressure study of $CeRhIn_5$ for magnetic field perpendicular to the ab-plane via specific heat measurements, where we show that superconducting characteristic parameters such as upper critical field H_{c2} and specific heat jump anomaly at T_c, are renormalized in the vicinity of the quantum critical point. We also argue that the new quantum phase with hidden magnetism in $CeRhIn_5$ may be directly connected to the spatially textured superconducting phase or Fulde–Ferrell–Larkin–Ovchinnikov (FFLO) state in $CeCoIn_5$.

2. Experiments

Single crystals of $CeRhIn_5$ are grown by indium flux [2]. The amount of free indium in the used crystal is less than that can be detected both by SQID and electrical resistivity. Those crystals show very high residual resistivity ratio, $R(300\text{ K})/R(1.1\text{ K})$, typically of order 440, indicating high quality. A hybrid Be–Cu/NiCrAl clamp-type pressure cell was used to achieve hydrostatic pressure up to 2.5 GPa and silicone fluid, as a transmitting medium. Pressure at low temperature was determined inductively by measuring the superconducting temperature of Sn manometer and sharp SC transition, which is typically <30 mK, ensures the hydrostaticity of the pressure cell. Specific heat was measured by ac calorimetry using a Stanford Research lock-in amplifier SR850, where Cr–Au/Fe(0.07%) thermocouple and constantan wires were used as a thermometer and heater, respectively [9]. In order to take into account the magnetic field dependence of the thermocouple wire, non-magnetic compound was measured under field and used to normalize the in-field data of $CeRhIn_5$.

3. Results and discussion

Fig. 1a shows the specific heat of $CeRhIn_5$ as a function of temperature at $P = 1.6$ GPa, representing C_p for pressures below P_1, the coexisting state. A λ-like jump due to the antiferromagnetic transition with $\mathbf{Q} = (0.5,0.5,0.297)$ occurs at 2.35 K, which is followed by the superconducting transition at a lower temperature (=1.5 K). Similar to $H \| ab$-plane, magnetic field applied perpendicular to the ab-plane suppresses T_c and slightly increases T_N (see Fig. 1b). At this field direction, however, there is no spin reorientation transition as was observed in $H \| ab$-plane. At 33 kOe, superconductivity is completely suppressed, but T_N is increased only by 50 mK compared to 0 kOe, making this field an ideal reference to subtract magnetic contribution to C_p. Inset to Fig. 1a shows the zero-field specific heat after subtracting magnetic background, i.e., $\Delta C = C(0\text{ kOe}) - C(33\text{ kOe})$. The superconducting discontinuity ratio $\Delta C_{SC}/C_N$ is 0.2, which is anomalously small compared to the BCS value (=1.43). Here C_N is the normal state value at T_c. Another characteristic superconducting parameter H_{c2} also displays abnormal temperature dependence (see Fig. 1b). Rather than showing characteristic saturation at low temperatures, H_{c2} shows linear T-dependence down to the lowest experimental temperature ($\sim 0.2T_c$) [3].

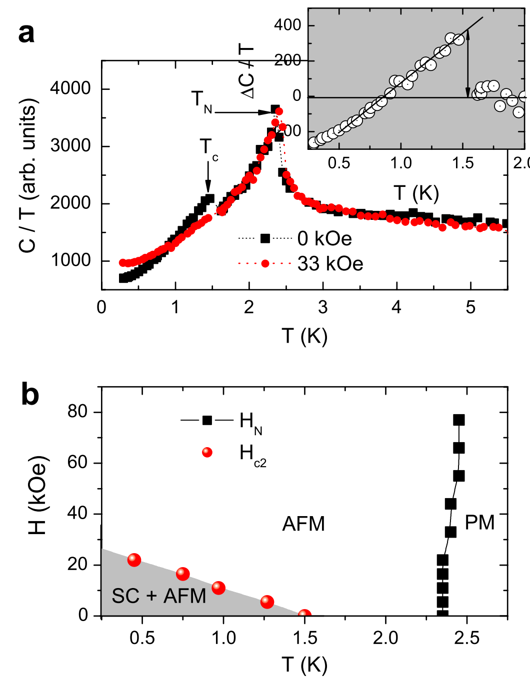

Fig. 1. (a) C/T versus T for $H = 0$ (squares) and 33 kOe (circles) at 1.6 GPa. T_c and T_N represent superconducting and antiferromagnetic transition temperatures, respectively. Inset: $\Delta C/T$ versus T, where $\Delta C = C(0\text{ kOe}) - C(33\text{ kOe})$. Solid lines are guides to eyes. (b) H–T phase diagram at 1.6 GPa. PM and AFM indicates paramagnetic and antiferromagnetic state.

The small jump of the specific heat at T_c has been also reported in heavy-fermion compounds $CePt_3Si$, URhGe etc., where antiferromagnetic transition occurs first at higher temperature and SC transition follows at a lower temperature [10]. Considering microscopic coexistence of magnetism and superconductivity below T_c in $CeRhIn_5$, we may postulate that part of the Ce 4f-electrons participate in RKKY-type magnetic ordering while the rest condensates at T_c, thus giving rise to the small jump. The ratio between the localized magnetic electrons and the delocalized SC electrons could be tuned by control parameters such as magnetic field, chemical substitution, or pressure. Indeed, the jump ratio increases to 2 (see Fig. 2a) for $P > P_1$ in $CeRhIn_5$, where the antiferromagnetism is completely suppressed. On the contrary to pressure effects, Cd doping to $CeCoIn_5$ indium site induces magnetism [11] and reduces the jump ratio from 4.5 at $x = 0$ to 0.3 at $x = 0.1$. Even though the small SC jump in C_p in the

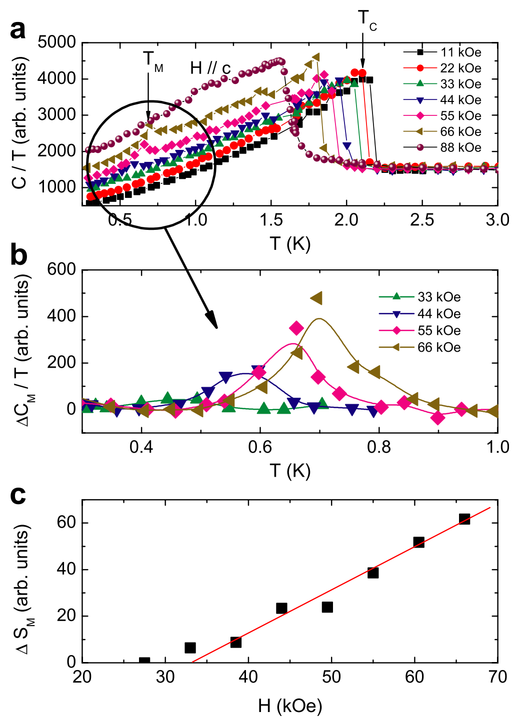

Fig. 2. (a) C/T versus T at 1.95 GPa for $H\perp ab$-plane. (b) $\Delta C\_M/T$ after subtracting smooth varying superconducting background. (c) Entropy involved in the field-induced magnetism plotted as a function of field H. Solid line is a guide to eyes.

coexisting phase of magnetism and superconductivity suggests that only fraction of the Fermi surface forms Cooper pairs at T_c, the observation of small jump in $CeIrIn_5$ in pure superconducting state indicates that this scenario has to be further studied to prove its validity conclusively.

Fig. 2a shows the specific heat of $CeRhIn_5$ at 1.95 GPa for $H\perp ab$-plane. At low fields up to 22 kOe, only superconductivity is observed. At 33 kOe, however, a weak anomaly starts to appear at 0.45 K, which is well below the superconducting transition temperature 2.08 K. When $H\|ab$-plane, similar anomaly appears at 11 kOe and 0.4 K at $P = 1.9$ GPa. Fig. 2b blows up the field-induced feature after subtracting smoothly varying background. The low-T feature is enhanced with increasing field and the characteristic temperature T_M increases, suggesting that this feature is intrinsic, not due to extrinsic effects. Similar to $H\|ab$-plane, we postulate that this low-T specific heat anomaly is due to a long-range antiferromagnetic order in the mixed superconducting state, possibly with the same **Q** for $P < P_1$. Entropy (ΔS_M) involved in this H-induced magnetism is calculated from Fig. 2b and is plotted against H in Fig. 2c. The entropy ΔS_M, an approximate measure of the effective magnetic moment, linearly depends on H. Since the areal density of the flux lattice or vortices is proportional to H, the vortex cores, normal region in superconductors, seems playing a crucial role in forming the magnetic ordering state.

Recent neutron scattering measurements revealed magnetism both in hole-doped and electron-doped high-T_c cuprates in the mixed superconducting state [12]. Since the size of the vortex cores is much smaller than the magnetic correlation length, it was argued that the magnetism should be delocalized outside vortex cores, coexisting with superconductivity. Demler et al. assumed that the cuprates are very close to a bulk quantum phase transition to a microscopic coexisting state of superconducting and spin density wave (SDW) orders [13]. When repulsive coupling between SC and SDW orders are considered, it is shown that delocalized magnetic state outside the vortex cores are preferred over the state confined within the cores. In our previous report, we have shown that the quantum phase transition in $CeRhIn_5$ follows the prediction from the theory that is specifically designed for the high-T_c cuprates, suggesting that hidden magnetism observed in $CeRhIn_5$ may be deconfined outside the vortex cores as in the cuprates. Neutron scattering measurements of $CeRhIn_5$ under high pressure are in progress to determine the nature of the hidden magnetism.

Fig. 3a shows the specific heat of $CeRhIn_5$ at 2.06 GPa for $H\perp ab$-plane. Compared to 1.95 GPa, H-induced magnetism first starts to appear at much higher field (66 kOe), but shows similar evolution of T_M with further increasing field. The bulk superconducting transition temperature is 2.27 K, which is compatible to the highest T_c observed among heavy fermion compounds, i.e., 2.25 K in $CeCoIn_5$ at ambient pressure. With increasing field, T_c is suppressed at a initial slope of $dH_{c2}/dT \approx 220$ kOe, predicting $H_{c2}(0) = 350$ kOe. The specific heat jump at T_c, $\Delta C/C_N$, is 2.5 and decreases with increasing field as expected in

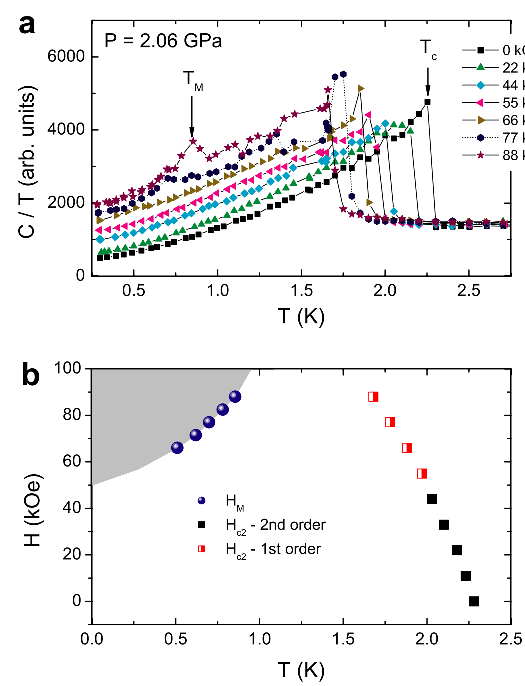

Fig. 3. (a) C/T versus T at 2.06 GPa for $H\perp ab$-plane. (b) H–T phase diagram based on specific heat measurements. H_M represents the critical field to induce magnetism in the mixed state. Filled (half-filled) squares display second-order (first-order) upper critical field H_{c2}.

typical type II superconductors. Above 55 kOe, surprisingly, the jump starts to increase and the peak shape becomes sharper and more symmetric, indicating that the nature of the superconducting transition has possibly changed from second to first order. Fig. 3b shows H–T phase diagram at 2.06 GPa. The change to the first order-like phase transition occurs almost at the same field where the low-T coexisting phase appears. At 1.95 GPa, however, H_M is 33 kOe while $H_{c2}^{(1st)}$ is 55 kOe, showing separation between the two characteristic fields.

The coexisting phase of SC and magnetic orders that carves out the low-T and high-H part of the superconducting phase in the pressurized $CeRhIn_5$ is reminiscent of the spatially inhomogeneous SC phase or FFLO phase in $CeCoIn_5$ [14]. Some of the similarities between the two compounds are as following: (i) magnetic field induces the new phases inside unconventional superconductivity, (ii) superconducting phase transition becomes first-order, (iii) entropy associated with the new phases linearly depends on the applied field. Recently, nuclear magnetic resonance (NMR) measurements of $CeCoIn_5$ argued that the supposed FFLO phase may not be conventional inhomogeneous superconducting state that consists of alternating normal and SC planes, but rather is relevant to some type of magnetism [15]. Even though the striking similarity between the two compounds and the new analysis on the nature of the FFLO phase suggest that magnetism plays a significant role both in hidden magnetic state in $CeRhIn_5$ and FFLO in $CeCoIn_5$, it is fair to note the dissimilarity: (i) the first-order SC transition occurs for $T < 0.5T_c$ in $CeCoIn_5$, while it is observed even at 0.85 T_c in $CeRhIn_5$, (ii) the FFLO phase boundary in $CeCoIn_5$ does not seem penetrating the H_{c2} line, but the H_M boundary goes through H_{c2} for $H \| ab$-plane in $CeRhIn_5$. With recent theoretical development that the first-order SC transition may not be relevant to the FFLO phase and the disappearance of T_M at 88 kOe and 1.95 GPa of $CeRhIn_5$ for $H \perp ab$-plane (see Fig. 2a), the opposing arguments (i) and (ii) against the magnetism scenario become moot, warranting more theoretical and experimental study on these new quantum phases.

4. Summary

We have reported pressure study of $CeRhIn_5$ for $H \perp ab$-plane. The small specific heat jump ratio at T_c is discussed in terms of the interplay between magnetism and superconductivity. Similar to$H \| ab$-plane, field-induced quantum phase transition to the coexisting phase of magnetism and superconductivity is identified. The first-order like SC transition for $P > P_1$ adds to the similarity between the hidden magnetism and the FFLO phase, warranting more theoretical and experimental study on these new quantum phases.

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