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Coexistence of antiferromagnetism and superconductivity in $CeRhIn_5$ under high pressure and magnetic field

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New light is shed on the interplay between antiferromagnetism and superconductivity by a detailed study of the phase diagram of $CeRhIn_5$ under high pressure and magnetic field using ac calorimetry. In zero field, the antiferromagnetic order vanishes suddenly above $p_c^\star=2$ GPa leading to a pure superconducting ground state. An inhomogeneous superconducting phase transition is found below $p_c^\star$. Above $p_c^\star$, the application of an external magnetic field $H\|ab$ induces a second transition, which suggests the reentrance of antiferromagnetism, its coexistence with superconductivity, and also the important role of the vortices. This field-induced supplementary state vanishes around the expected quantum critical point, which will occur in the absence of superconductivity.

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The discovery of the cerium heavy fermion $CeMIn_5$ (M=Co, Ir, Rh) systems opened a new route to investigate the interplay of magnetic fluctuations, unconventional superconductivity (SC) and antiferromagnetic order (AF) in strongly correlated electron systems in the vicinity of a quantum phase transition (QPT).[1–3] While $CeCoIn_5$ is an unconventional superconductor very close to a QPT at ambient pressure (p) with d-wave symmetry below $T_c=2.2$ K,[4–6] $CeRhIn_5$ offers the possibility to tune a heavy fermion system from AF to SC as a function of pressure. At $p=0$, $CeRhIn_5$ orders antiferromagnetically below the Néel temperature $T_N=3.8$ K in an incommensurate helical structure with propagation vector $Q=(1/2,1/2,0.297)$ and a staggered moment of about $0.8\mu_B$.[7] Neutron scattering experiments under pressure showed that this magnetic structure is almost unchanged up to 1.7 GPa.[8] SC has been reported for $p>0.9$ GPa on the basis of resistivity measurements.[8] Nuclear-quadrupole resonance (NQR) measurements have shown that SC and AF may coexist on a microscopic scale in the pressure range from $p=1.6$–1.75 GPa with a gapless nature of the low-lying excitations.[9] A pure superconducting ground state with line nodes in the gap is attained for $p>2$ GPa.[10] Previous detailed specific heat measurements under hydrostatic pressure give evidence that the magnetic transition disappears above $p_c^\star\approx1.95$ GPa.[11] Measurements of quantum oscillations show a change of the Fermi surface (FS) together with a strong increase of the effective mass of the carriers near 2.35 GPa suggesting a quantum critical point (QCP) in this pressure range.[12]

The application of an external magnetic field, H, offers a second parameter to influence the competition between AF and SC.[13] Recently, it was shown that a magnetic QCP can be achieved in $CeCoIn_5$ by the application of a magnetic field of the order of the upper critical field $H_{c2}(0)$.[14,15] Close to $H_{c2}(0)$ a new superconducting phase appears which is interpreted as a Fulde-Ferrel-Ovchinnikov-Larkin (FFLO) state.[16–20] However, this state may also originate from magnetic interactions.

The high magnetic field phase diagram of $CeRhIn_5$ at $p=0$ has been studied by specific heat,[21] magnetization,[22,23] and thermal expansion measurements.[24] For H in the ab plane, T_N increases slightly up to 3.9 K with an increasing field. The magnetization in the ab plane shows a small step-like increase near $H_m^\star=2$ T at low temperatures, due to a change in the magnetic structure most likely associated with a reorientation of the helix and/or a change from an incommensurate AF^I to a commensurate structure AF^{III}.[21] Consequently for $H>H_m^\star$, a second magnetic transition below T_N appears at $T_{N1}\approx3.5$ K. A transition to a spin-polarized paramagnetic state (PM) is observed for $H>H_m\approx50$ T.[23] For $H\|c$ no second phase under field appears.[21]

Here we report on ac calorimetry under pressure and magnetic field $H\|ab$ in $CeRhIn_5$.[25] The pressure cell used is the same as in our previous experiment.[11] The pressure is measured *in situ* by the ruby fluorescence method, the absolute pressure accuracy is better than 0.05 GPa. Heating is realized by a laser diode, tunable in frequency and power. The frequency was chosen to be slightly above the cutoff $\nu_c\approx400$ Hz at the lowest temperature whatever the pressure. The temperature oscillations ($T_{ac}<5$ mK) are measured with an AuFe/Au thermocouple which is soldered on the sample. The specific heat can be estimated by $C_{ac}\propto-S_{th}\sin\theta/V_{th}2\pi\nu$ where S_{th} corresponds to the thermoelectric power of the thermocouple, V_{th} and θ are the measured voltage and phase of the thermocouple signal. The average temperature of the sample has been measured with a calibrated Cernox sensor which is placed in the field compensated zone of a superconducting magnet. The ac calorimetry method has been verified at zero pressure under magnetic field. The measurements were performed in a ^{3}He cryostat ($T>0.5$ K) where a magnetic field of 7.5 T could be applied in the ab plane of the crystals.

Figure 1 shows C_{ac}/T in zero field in the pressure range from 1.2–2.17 GPa. In contrast to the previous measurements which had been limited to $T>1.5$ K,[11] in the new measurements two anomalies appear for $1.5<p<1.9$ GPa. The lower temperature transition, which corresponds to SC, is tiny for $p<p_c^\star\approx1.95$ GPa in agreement with NQR data.[9] However, we cannot conclude if a new homogeneous gapless SC is formed which coexists with AF (Ref. 26) or if the weak

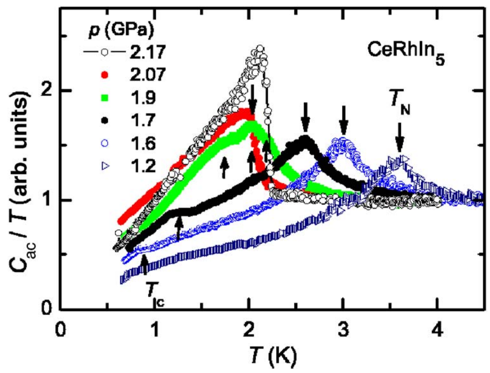

FIG. 1. (Color online) Temperature dependence of the ac specific heat divided by temperature for different pressures. Arrows indicate the superconducting transition temperatures T_c (↑) and the Néel temperature T_N (↓), respectively. Below $p_c^\star \approx 1.95$ GPa the superconducting anomaly is very small. For p slightly above $p_c^\star$ a nice superconducting anomaly appears. (Data are normalized to 1 at 4 K.)

SC anomaly at T_c reveals only a strong sensitivity to imperfections, for example due to a mismatch between incommensurate magnetic ordering (AF^I) and SC or due to additional nodes in the gap function of the SC state caused by the crossing of the FS with the magnetic Brillouin zone.[27] Direct evidences on the inhomogenous superconducting transition below $p_c^\star$ are given by the large discrepancy between T_c detected by resistivity T_c^ρ,[8] ac susceptibility T_c^χ,[11] and the present specific heat measurements T_c^C with a sequence $T_c^\rho > T_c^\chi > T_c^C$. Up to 1.5 GPa, the resistivity anomaly at T_c^ρ may not be a bulk property; a similar case is reported for $CeIrIn_5$.[3,28] By contrast, the specific heat measurement clearly shows a sharp and very large superconducting anomaly at $p=2.17$ GPa indicating a pure superconducting ground state, basically gapped. At $p=2.07$ GPa no extra AF transition can be detected below T_c. Above $p_c^\star$, an eventual domain of a coexistence of AF and SC will be extremely narrow in pressure and experimentally very difficult to point out.

The (p,T) phase diagram of $CeRhIn_5$ in zero field is summarized in Fig. 2 (data from Ref. 11 have been included). The low temperature specific heat measurements clearly show the interplay of AF and SC in the pressure range from 1.6–1.9 GPa. A first order transition seems to emerge at $p_c^\star = 1.95$ GPa, where $T_c \approx T_N$. A linear p extrapolation of T_N to zero temperature indicates that T_N may be fully suppressed near the pressure of the maximum of T_c at $p_c \approx 2.4 \pm 0.1$ GPa and the maxima of some of the effective masses.[12] If $T_c > T_N$, the ground state in zero field seems purely superconducting with d-wave symmetry[10] as in $CeCoIn_5$.[4,5] The opening of a superconducting gap on large parts of the main FS leads to the suppression of the magnetic ordering.

Figure 3 shows the specific heat for various fields $H \| ab$ at different pressures below and above $p_c^\star$. The (H,T) phase diagrams obtained from these data are displayed in Fig. 4. At 1.2 GPa, in the normal AF state, three magnetic transitions appear for $H > 3$ T. By comparison to $p=0$ results[21] the magnetic phase diagram is only weakly p dependent [see Fig.

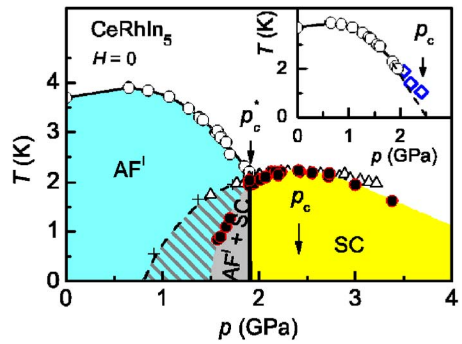

FIG. 2. (Color online) (p,T) phase diagram of $CeRhIn_5$ from specific heat (○, ●), susceptibility (△, Ref. 11), and resistivity measurements (×, Ref. 8). Below 1.5 GPa $CeRhIn_5$ orders in an incommensurable structure AF^I, the hatched area indicates an inhomogeneous superconducting state, and AF^I+SC corresponds to the region where SC appears in the specific heat experiment in the magnetically ordered state below T_N. A pure superconducting state SC is realized above $p_c^\star$. The vertical line marks a possible first order transition from AF^I+SC to SC. The inset shows the extrapolation of T_N to zero in the absence of SC. (◇) indicates the temperature where $T_M(H)$ crosses $T_c(H)$, and corresponds to T_N if SC is suppressed (see also Fig. 4).

4(a)]. For $p=2.07$ GPa the superconducting transition at 0 T is still rather broad, but above H_{c2} another phase transition at T_M appears in the normal state [Fig. 4(b)]. At 2.2 GPa the transition has a width less than 0.1 K at $H=0$. For $H > 4$ T, the additional transition develops on cooling already inside the superconducting phase and survives entering in the normal phase. No second transition can be detected below $H \approx 4$ T. For $H \geq 4$ T two well separated anomalies inside the superconducting state become obvious at 2.41 GPa, almost as observed in UPt_3.[29] For higher pressures ($p=2.73$ GPa), only a unique superconducting phase persists.

The assignment of the superconducting anomaly under magnetic field is unambiguously given by the form of the $H_{c2}(T)$ curve, also in comparison to previous resistivity data.[30] There are strong indications that the observed second anomaly is associated to AF. This transition at T_M is almost H independent, at least for $H > 4$ T, as the antiferromagnetic transition at $p=0$ and $p=1.2$ GPa. The inset of Fig. 2 shows the p dependence of the crossing temperature corresponding to $T_c(H) \sim T_M(H)$ for $p > p_c^\star$. Its extrapolation to zero is obtained for $p \approx 2.6$ GPa, slightly higher than p_c. That can indicate the enhancement of T_N when AF and SC coexist in this mixed state. In the simplest model, weak coupling in a clean limit, the p dependence of the effective mass $m^\star$ of the quasiparticles can be estimated from the initial slope $H'_{c2} = dH_{c2}/dT \propto (m^\star)^2 T_c$.[32] For $p=2.07$ GPa we find $H'_{c2} = 20$ T/K, near 2.41 GPa H'_{c2} increases to 33 T/K and decreases to 25 T/K for 2.73 GPa while T_c is almost unchanged.[33] This, as well as the size of the specific heat jump at T_c, indicates that $m^\star$ has its maximum near 2.4 GPa in agreement with the expected variation of $m^\star$ at p_c for an antiferromagnetic QCP.[35]

The new phase presumably with AF and SC appears for $p > p_c^\star$ only above some critical field of the order $H \approx 4$ T.[31]

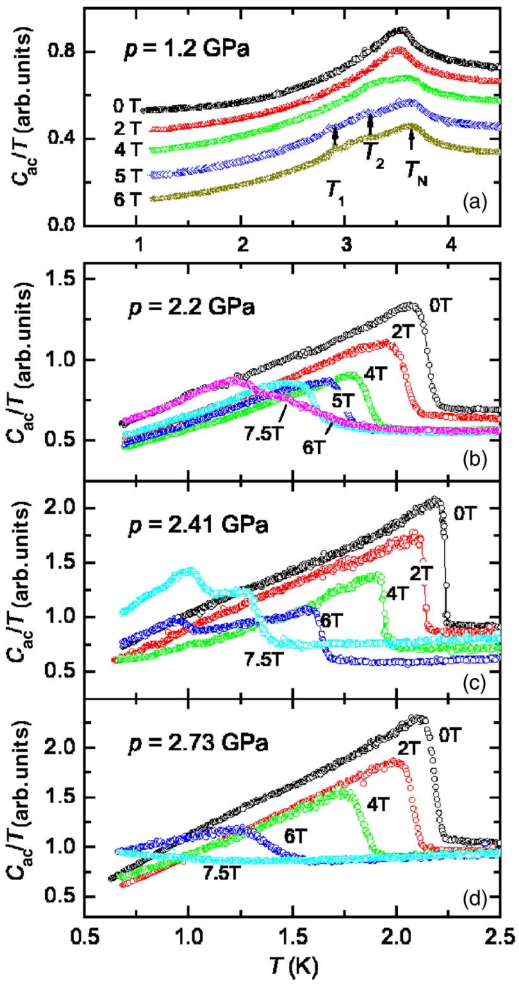

FIG. 3. (Color online) C_{ac}/T vs the temperature of $CeRhIn_5$ at different pressures for various magnetic fields $H \| ab$. [Data in (a) are shifted for clarity.]

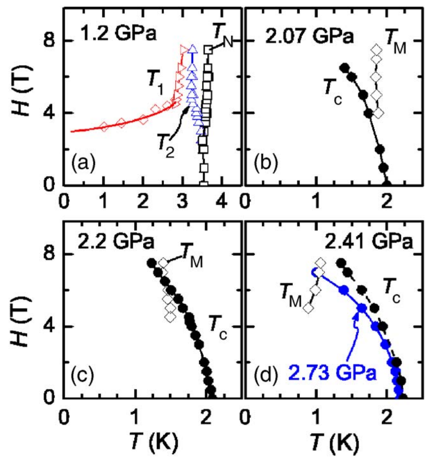

FIG. 4. (Color online) (H,T) phase diagram of $CeRhIn_5$: (a) below $p_c^\star$ at 1.2 GPa; (b)–(d) above $p_c^\star$ at 2.07, 2.2, 2.41, and 2.73 GPa. (●) correspond to the upper critical field H_{c2}, and (◇) to the field induced magnetic transition at T_M, respectively. Lines are to guide the eyes.

At 2.2 and 2.41 GPa its regime expands far below the field where $T_c(H)$ and $T_M(H)$ cross. In the superconducting domain below H_{c2}, AF can originate from the vortex core (see for cuprates Refs. 36 and 37). Interestingly, the lowest field where the second phase is detected above $p_c^\star$ is close to the characteristic field $H_m^\star$ where the magnetic structure changes for $H \| ab$ at $p=0$, which is, as indicated above, weakly pressure dependent. Gapped SC may coexist probably only with the AF^{III} phase above $p_c^\star$. However, detailed neutron scattering or NMR experiments are indispensable to resolve the nature of the new field induced phase.

In conclusion, the (p,T,H) phase diagram of $CeRhIn_5$ is extremely rich. Up to 1.5 GPa the ground state is antiferromagnetic AF^I in zero field and the magnetic properties are almost unchanged in comparison to $p=0$ (see also Ref. 8). A modification of the magnetic structure appears for $H_m^\star \gtrsim 2$ T from AF^I to AF^{III}. The saturation field H_m is surely one order of magnitude higher than this field for p slightly below p_c.[38] From 1.5–1.95 GPa the ground state is AF+SC for $H=0$, with an inhomogeneous SC phase transition. Above $p_c^\star \approx 1.95$ GPa, when $T_N(p)$ will cross $T_c(p)$ an almost fully gapped SC state expels the AF state. The new superconducting state of the low temperature-high magnetic field phase may result from the unique situation that both, the magnetic and superconducting coherence lengths related, respectively, to the magnetic QPT and clean unconventional SC have very close values. The main difference to the results published in Ref. 25 is that we do not observe the field induced phase for fields below $H \approx H_m^\star$. In Ref. 25 the boundary between the pure SC state and the induced AF+SC state is reported to increase linearly in magnetic field in agreement with the model in Ref. 37. However, the magnetic transition from AF^I to AF^{III} or a critical vortex density may have a strong impact on the appearance of the field induced state at least on the shape of the superconducting anomalies. The specific heat anomaly of the field reentrant phase are certainly different below and above 4 T. In the high field domain ($H > 4$ T), this reentrance appears related with uniform AF; below this field it may originate from the nucleation of antiferromagnetism in the vortex core in agreement with the SO(5) theory.[37] However, the precise boundary close to $p_c^\star$ as well as microscopic investigations of the magnetic state under field deserve a new set of experiments.[39]

We want to emphasize that a first order separation between AF and clean SC or at least a fast drop of T_N before T_c reaches its maximum value may be a quite general phenomenon at least for heavy fermion superconductors. In zero field, the condition $T_N > T_c$ for these highly correlated systems may lead to the impeachment to approach the magnetic QCP, i.e., a second order QPT where T_N decreases continuously to zero at p_c for $H=0$. Fortunately for $CeRhIn_5$ the maxima of T_N and T_c have quite comparable values. In other, more three dimensional heavy fermion compounds like $CeRh_2Si_2$, $CePd_2Si_2$, or $CeIn_3$ the maximum of T_N is about ten times higher than T_c and the domain of competition between AF and SC is difficult to observe as it occurs in a narrow temperature and field range.[38] For $CeIn_3$ indications for the first order nature of the transition between AF and SC has been shown by NQR measurements.[41] Further, the repulsion between AF and SC was one of the important issues of

$CeCu_2Si_2$ for decades (see, e.g., Refs. [42,43]). One can speculate if the QCP exists, i.e., a second order QPT in the presence of SC. Finally, let us point out the drastic difference between heavy fermion superconductors close to a magnetic instability and conventional magnetic superconductors as Chevrel phases with two separated classes of electrons (localized and delocalized) where AF is very robust and can appear well below T_c.[44]

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