Gapless Magnetic and Quasiparticle Excitations due to the Coexistence of Antiferromagnetism and Superconductivity in $CeRhIn_5$: A Study of ^{115}In NQR under Pressure

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We report systematic measurements of ac susceptibility, nuclear-quadrupole-resonance spectrum, and nuclear-spin-lattice-relaxation time (T_1) on the pressure (P)-induced heavy-fermion superconductor $CeRhIn_5$. The temperature (T) dependence of $1/T_1$ at $P = 1.6$ GPa has revealed that antiferromagnetism (AFM) and superconductivity (SC) coexist microscopically, exhibiting the respective transition at $T_N = 2.8$ K and $T_c^{MF} = 0.9$ K. It is demonstrated that SC does not yield any trace of gap opening in low-lying excitations below $T_c^{onset} = 2$ K, but $T_c^{MF} = 0.9$ K, followed by a $T_1T =$ const law. These results point to the unconventional characteristics of SC coexisting with AFM. We highlight that both of the results deserve theoretical work on the gapless nature in the low-lying excitation spectrum due to the coexistence of AFM and SC and the lack of the mean-field regime below $T_c^{onset} = 2$ K.

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In recent years, an intimate interplay between antiferromagnetism (AFM) and superconductivity (SC) has been the most interesting and outstanding issue in cerium (Ce)-based heavy-fermion (HF) systems. The finding of P-induced SC in $CeCu_2Ge_2$ [1], $CeIn_3$, $CePd_2Si_2$ [2,3], and $CeRhIn_5$ [4] strongly suggest that AFM and SC are related to each other since P-induced SC occurs either when AFM vanishes or coexists with it [5]. Among them, the study on the cubic $CeIn_3$ and the quasi-two-dimensional tetragonal $CeRhIn_5$ is promising for systematic investigation of an intimate interplay between AFM and SC. A new HF antiferromagnetic compound $CeRhIn_5$ revealed an antiferromagnetic to superconducting transition at a relatively lower critical pressure $P_c = 1.63$ GPa and higher $T_c = 2.2$ K than in previous examples [1–4].

At $P = 0$, a previous study of ^{115}In NQR (nuclear-quadrupole-resonance) found that $CeRhIn_5$ has an incommensurate wave vector $\mathbf{q}_M = (1/2, 1/2, 0.297)$ below the Néel temperature $T_N = 3.8$ K [6]. A neutron experiment revealed the reduced Ce magnetic moments $M_s \sim 0.8\mu_B$ in the antiferromagnetic ordered state [7,8]. Our previous NQR study showed that T_N gradually increases up to 4 K as P increases up to $P = 1.0$ GPa and decreases with further increasing P [5,9,10]. In addition, the temperature (T) dependence of nuclear-spin-lattice-relaxation rate $1/T_1$ has probed pseudogap behavior at $P = 1.23$ and 1.6 GPa [10]. This suggests that $CeRhIn_5$ may resemble other strongly correlated electron systems [11,12]. At $P =$ 2.1 GPa above P_c, $1/T_1$ decreases, obeying a T^3 law without the coherence peak just below T_c. This indicates that the SC of $CeRhIn_5$ is an unconventional one and it has line-node gap [9,13]. Most remarkably, at $P = 1.75$ GPa, the onset of AFM is evidenced from a clear split in ^{115}In NQR spectrum due to the spontaneous internal field H_{int} below $T_N = 2.5$ K. Simultaneously, bulk SC below $T_c =$ 2.0 K is demonstrated by the observation of the Meissner diamagnetism signal. These results have demonstrated that AFM coexists homogeneously with the SC at a microscopic level at the border where both SC and AFM meet one another [5].

In this Letter we report systematic measurements of ac susceptibility (χ_{ac}) in $P = 1.12$–2.0 GPa and focus on novel superconducting characteristics in the coexistent state of AFM and SC for $CeRhIn_5$ at $P = 1.6$ GPa.

$CeRhIn_5$ forms in $HoCoGa_5$-type structure where $CeIn_3$ and $RhIn_2$ layers alternately stack along the c axis. Accordingly, there are two inequivalent In sites per unit cell for NQR measurements. The ^{115}In NQR measurements at the In(1) site surrounded by four Ce atoms were performed using a laboratory-built spectrometer [5,6,9,10]. The NQR spectrum was obtained by plotting the intensity of a spin-echo signal as a function of frequency at $1\nu_Q$ ($\pm 1/2 \leftrightarrow \pm 3/2$) transition which is the lowest one of the four transitions for the In nuclear spin $I = 9/2$. The ^{115}In NQR T_1 was measured at the transitions of $2\nu_Q$ ($\pm 3/2 \leftrightarrow \pm 5/2$) above $T = 1.4$ K, and at $1\nu_Q$ below $T = 1.4$ K with the saturation-recovery method. The high-frequency χ_{ac} measurements under P were carried out by measuring the inductance of an *in situ* NQR coil as reported in the previous report [9]. To obtain hydrostatic P, a BeCu piston-cylinder cell was used with Daphne oil (7373) as a P-transmitting medium up to $P =$ 2.0 GPa. For our P cell, a value of P distribution ($\Delta P/P$) is less than 3% at low T, determined from a broader NQR linewidth at $P = 1.6$ GPa than at ambient P.

Figure 1 indicates a rich P-T phase diagram of $CeRhIn_5$ for AFM and SC referred to in the previous report [5]. The SC seems to survive under AFM near the magnetic criticality in $CeRhIn_5$. Note that the present measurement of χ_{ac} reveals progressive reduction in the value of bulk

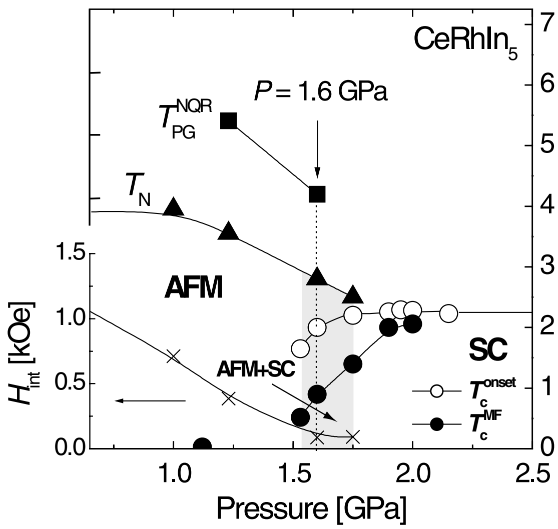

FIG. 1. The P-T phase diagram for $CeRhIn_5$. The respective marks denoted by squares, triangles, and crosses correspond to the pseudogap temperature T_{PG}^{NQR}, the antiferromagnetic ordering temperature T_N, and the internal field H_{int} at the In site. The open and solid circles correspond to the onset temperature T_c^{onset} and T_c^{MF} of superconducting transition (see text). The dotted line denotes the position for $P = 1.6$ GPa. The shaded region indicates the coexistent P region of AFM and SC.

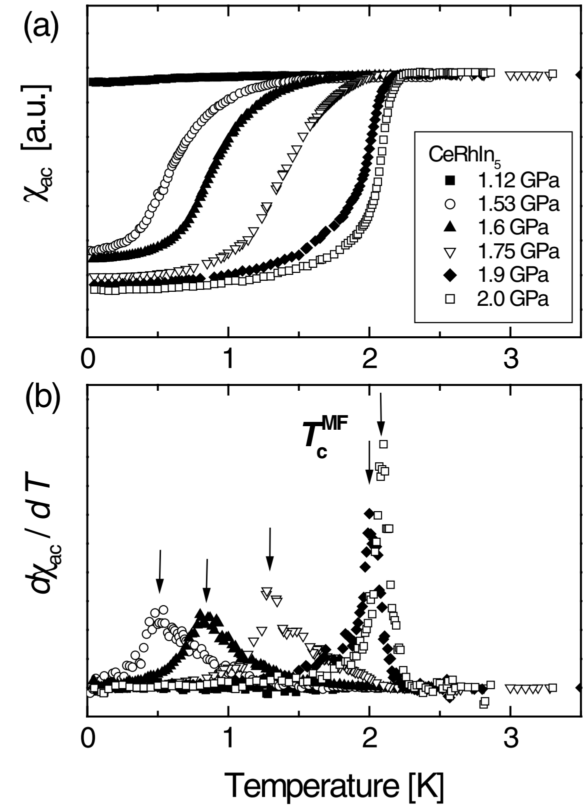

FIG. 2. (a) The T dependencies of χ_{ac} and (b) the T derivatives of χ_{ac} ($d\chi_{ac}/dT$) at $P = 1.12$–2.0 GPa. The arrows indicate T_c^{MF} at each value of P (see text).

superconducting transition temperature T_c^{MF}, as shown by closed circles in Fig. 1.

Figures 2(a) and 2(b) indicate the respective T dependencies of χ_{ac}'s and their T derivatives $d\chi_{ac}/dT$ in $P = 1.12$–2.0 GPa. At $P = 1.12$ GPa, χ_{ac} decreases slightly below 1 K with a size in reduction of χ_{ac} ($\Delta\chi_{ac}$) due to SC being about only 4% of $\Delta\chi_{ac}$ (bulk) for the bulk Meissner diamagnetism at $P = 2.0$ GPa. It remains unclear whether this reduction arises from some diamagnetism associated with superconducting fluctuations or possible experimental uncertainty. At least it may be ruled out that bulk superconducting transition does not occur at $P = 1.12$ GPa. We note that the specific-heat result under P suggests P_c(SC) ~ 1.5 GPa at which the bulk SC sets in [14]. Consistently, a bulk nature of SC in a range $P = 1.53$–2.0 GPa is corroborated by the observation of the Meissner diamagnetism signal whose size is almost the same as in the exclusively superconducting phase. Thus, the SC in $CeRhIn_5$ emerges at pressures exceeding P_c(SC) ~ 1.5 GPa, as seen in Fig. 1. As seen in Fig. 2(a), however, it should be noted that the superconducting transition width becomes significantly broader with decreasing P. A similar behavior is also seen in the P dependence of resistivity over the same P range. Although a zero resistance is observed at T_c^{zero}, the decrease in resistance towards T_c^{zero} becomes broadest at $P = 1.6$ GPa [see Fig. 5(b)] [4]. $\Delta\chi_{ac}$'s at $P = 1.53$ and 1.6 GPa decrease to 80% of $\Delta\chi_{ac}$ (bulk) for the bulk Meissner diamagnetism at $P = 2.0$ GPa. These anomalous phenomena are associated with novel superconducting characteristics inherent to the microscopic coexisting state of AFM and SC, but not due to some phase separation between SC and AFM. This is because the homogeneous AFM over the whole sample is evidenced at $P = 1.6$ GPa without any trace for the phase separation between AFM and SC, as shown below. As seen in Fig. 2(a), T_c^{onset} is defined as a temperature below which the diamagnetism starts to appear, whereas a peak of $d\chi_{ac}/dT$ as T_c^{MF}, as seen in Fig. 2(b).

Next we present microscopic evidence for novel superconducting characteristics at the coexisting state of AFM and SC in $CeRhIn_5$ at $P = 1.6$ GPa. The inset of Fig. 3 displays the NQR spectra above and below T_N at $P = 1.6$ GPa. Below $T_N = 2.8$ K, the NQR spectrum splits into two peaks due to the appearance of H_{int} at the In site. This is clear evidence for the occurrence of AFM at $P = 1.6$ GPa as well as reported in the previous result at $P = 1.75$ GPa [5]. The plots of $H_{int}(T)/H_{int}(0) = M_s(T)/M_s(0)$ vs (T/T_N) at $P = 0$ and 1.6 GPa are compared in Fig. 3, showing nearly the same behavior. Here $H_{int}(0)$ is the value extrapolated to $T = 0$ K and $M_s(T)$ is the T dependence of spontaneous staggered magnetic moment. The character of AFM at $P = 1.6$ GPa is expected to be not so much different from that at $P = 0$.

Figure 4 indicates the T dependence of $1/T_1$ at $P = 1.6$ GPa. A clear peak in $1/T_1$ is due to critical magnetic fluctuations at $T_N = 2.8$ K. Below $T_N = 2.8$ K, $1/T_1$ continues to decrease moderately down to $T_c^{MF} = 0.9$ K

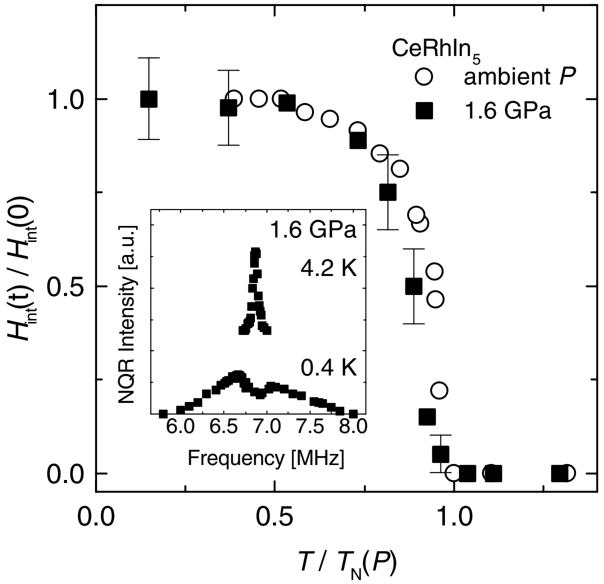

FIG. 3. Plots of $H_{\rm int}(T)/H_{\rm int}(0)$ vs T/T_N at $P = 0$ and 1.6 GPa (see text). The inset shows the ^{115}In NQR spectra of $1\nu_Q$ at $P =$ 1.6 GPa above and below $T_N = 2.8$ K.

even though passing across $T_c^{\rm onset} \sim 2$ K. This relaxation behavior suggests that the SC does not develop following the mean-field approximation below $T_c^{\rm onset}$. Most remarkably, $1/T_1$ decreases below $T_c^{\rm MF}$, exhibiting a faint T^3 behavior in a narrow T range. With further decreasing T, $1/T_1$ becomes proportional to the temperature, indicative of a gapless nature in low-lying excitation spectrum in the microscopically coexisting state of SC and AFM.

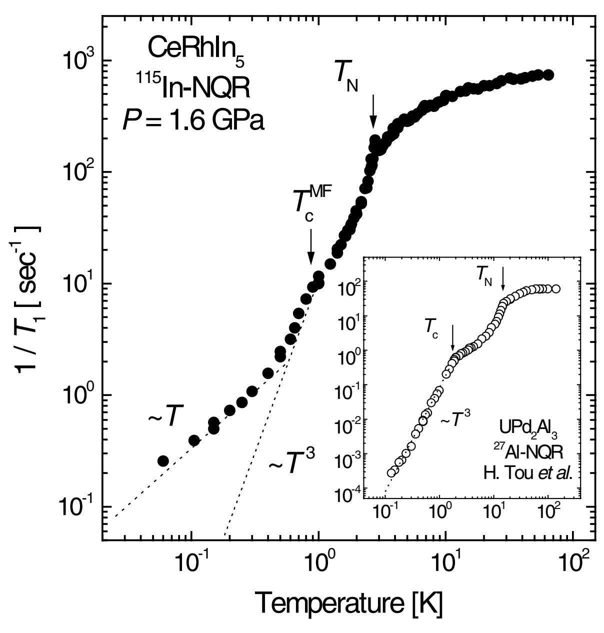

FIG. 4. The T dependence of $1/T_1$ at $P = 1.6$ GPa. Both dotted lines correspond to $1/T_1 \propto T$ and $1/T_1 \propto T^3$. The inset indicates the T dependence of ^{27}Al NQR $1/T_1$ of UPd_2Al_3 cited from the literature [15]. The dotted line corresponds to $1/T_1 \propto T^3$.

Thus the T_1 measurement unravels that an intimate interplay between AFM and SC gives rise to some amplitude fluctuations of superconducting order parameter between $T_c^{\rm onset}$ and $T_c^{\rm MF}$. Such fluctuations may be responsible for the broad transition in resistance and $\chi_{\rm ac}$ measurements and for the slightly reduced value less than $\Delta\chi_{\rm ac}$ (bulk) in the P range higher than $P = 1.75$ GPa (see Fig. 2). Furthermore, the $T_1T =$ const behavior well below $T_c^{\rm MF}$ evidences the gapless nature in the coexisting state of AFM and SC. This result is consistent with those in $CeCu_2Si_2$ at the border of AFM [16] and a series of $CeCu_2(Si_{1-x}Ge_2)_2$ compounds that show the coexistence of AFM and SC [17,18]. The specific-heat result that probed a finite value of its T-linear contribution, $\gamma_0 \sim$ 100 mJ/mol K^2 at $P = 1.65$ GPa, is now understood though not due to a first-order-like transition to SC [14], but to the gapless nature in the coexisting state of AFM and SC. It is noteworthy that such $T_1T =$ const behavior is not observed below T_c at $P = 2.1$ GPa [9], consistent with the specific-heat result under P as well [14]. This means that the origin for the $T_1T =$ const behavior below $T_c^{\rm MF}$ at $P = 1.6$ GPa is not associated with some impurity effect. If it were the case, the residual density of states below T_c should not depend on P. This novel feature differs from the uranium (U)-based HF antiferromagnetic superconductor UPd_2Al_3 which has multiple $5f$ electrons. In UPd_2Al_3, a superconducting transition occurs at $T_c = 1.8$ K, well below the long-range antiferromagnetic order at $T_N = 14.3$ K [19,20]. The ^{27}Al NQR T_1 results in UPd_2Al_3 are indicated in the inset of Fig. 4 [15]. $1/T_1$ decreases obeying a T^3 law over 3 orders of magnitude below the onset of T_c without any trace for $1/T_1T =$ const behavior. This is consistent with the line-node gap even in the coexisting state of AFM and SC below T_c.

In order to highlight the novel superconducting nature in a microscopic level, the T dependence of $1/T_1T$ is shown in Fig. 5(a) at $P = 1.6$ GPa in $T = 0.05$–6 K and is compared with the T dependence of the resistance $R(T)$ at $P = 1.63$ GPa referred to in the literature [4]. Although each value of P is not exactly the same, they only differ by 2%. We remark that the T dependence of $1/T_1T$ points to the pseudogap behavior around $T_{\rm PG}^{\rm NQR} =$ 4.2 K, the AFM at $T_N = 2.8$ K, and the SC at $T_c^{\rm MF} =$ 0.9 K at which $d\chi_{\rm ac}/dT$ has a peak, as seen in Fig. 5(b). This result itself evidences the microscopic coexistent state of AFM and SC. A comparison of $1/T_1T$ with the $R(T)$ at $P = 1.63$ GPa in Fig. 5(b) is informative in shedding light on the uniqueness of superconducting and magnetic characteristics. Below $T_{\rm PG}^{\rm NQR}$, $R(T)$ starts to decrease more rapidly than a T-linear variation extrapolated from the high T side. It continues to decrease across $T_N = 2.8$ K, reaching zero resistance at $T_c^{\rm zero} \sim 1.5$ K. The resistive transition width for SC becomes broader. Unexpectedly, $T_c^{\rm onset} \sim 2$ K, which is defined as the temperature below which the diamagnetism starts to appear, is higher than $T_c^{\rm zero} \sim 1.5$ K. Any signature for the onset of SC from the $1/T_1$ measurement is not evident between

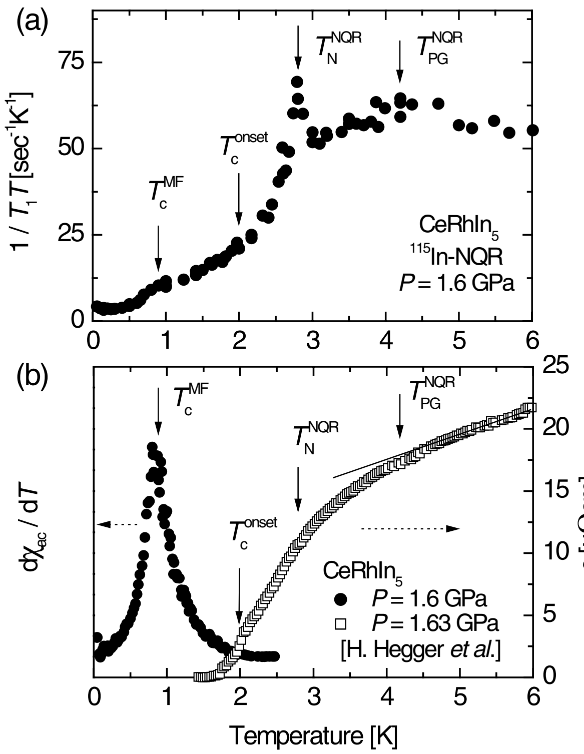

FIG. 5. (a) The T dependence of $1/T_1T$ at $P = 1.6$ GPa. (b) The T dependencies of $d\chi_{ac}/dT$ at $P = 1.6$ GPa and resistance at $P = 1.63$ GPa cited from the literature [4]. T_c^{MF} and T_c^{onset} correspond to the respective temperatures at which $d\chi_{ac}/dT$ has a peak and below which χ_{ac} starts to decrease. T_N corresponds to the antiferromagnetic ordering temperature at which $1/T_1T$ exhibits a peak and T_{PG}^{NQR} to the pseudogap temperature below which it starts to decrease. The solid line is guide for the eye for the T-linear variation in resistance at temperatures higher than T_{PG}^{NQR}.

T_c^{onset} and T_c^{MF}, demonstrating that the mean-field–type of gap does not grow up down to $T_c^{MF} \sim 0.9$ K. The existence of fluctuations due to the interplay of AFM and SC is responsible for the broad transition toward SC that coexists with AFM.

Finally, we remark that the recent neutron-diffraction experiment suggests almost the P-independent size of staggered moment M_s in the antiferromagnetic ordered state [8], in contrast to the large P dependence of H_{int}, as seen in Fig. 1. Its relatively large size of moment with $M_s \sim 0.8\mu_B$ seems to support such a picture that the same f electron exhibits simultaneously itinerant and localized dual nature because there is only one $4f$ electron per Ce^{3+} ion. In this context, it is natural to consider that the superconducting nature in the coexisting state of AFM and SC belongs to a novel class of phase which differs from the conventional d-wave SC with the line-node gap. As a matter of fact, a theoretical model has recently been put forth to address the underlying issue in the coexistent state of AFM and SC [21].

In conclusion, we have reported the microscopic coexistence of AFM and SC from the systematic measurements of the T dependencies of $1/T_1$ and NQR spectra for $CeRhIn_5$ at $P = 1.6$ GPa. Also, the coexistent state is suggested to persist down to $P \sim 1.5$ GPa, at least from the measurement of χ_{ac}. The detailed measurement of $1/T_1$ has revealed that SC does not yield any trace of gap opening in low-lying excitations below the onset temperature $T_c^{onset} = 2$ K, but $T_c^{MF} = 0.9$ K, followed by the $1/T_1T$ = const law in the low T regime. These results differ from any previous examples, pointing to the novel characteristics of SC coexisting with AFM. We highlight that both of the results deserve theoretical work on the gapless nature in low-lying excitation spectrum due to the coexistence of AFM and SC and the lack of the mean-field regime below $T_c^{onset} = 2$ K.

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