Coexistence of Antiferromagnetism and Superconductivity near the Quantum Criticality of the Heavy-Fermion Compound CeRhIn$_5$

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We report a study on the interplay between antiferromagnetism (AFM) and superconductivity (SC) in a heavy-fermion compound CeRhIn$_5$ under pressure $P = 1.75$ GPa. The onset of the magnetic order is evidenced from a clear split of ^{115}In nuclear quadrupole resonance spectrum due to the spontaneous internal field below the Néel temperature $T_N = 2.5$ K. Simultaneously, bulk SC below $T_c = 2.0$ K is demonstrated by the observation of the Meissner diamagnetism signal whose size is the same as in the exclusively superconducting phase. These results indicate that the AFM coexists homogeneously with the SC at a microscopic level.

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Understanding the interplay between magnetism and superconductivity (SC) is one of the central tasks in condensed-matter physics. In many strongly correlated electron systems such as high superconducting transition-temperature (T_c) copper oxides, low-dimensional organic salts, and heavy-mass electron (heavy-fermion) intermetallic compounds, the magnetism and SC are closely related. In some heavy-fermion (HF) systems, the relationship between the two phenomena are complicated; antiferromagnetically ordered and superconducting phases appear to coexist in some cases, which has been reported particularly in some uranium(U)-based HF systems. For example, UPd$_2$Al$_3$ exhibits a superconducting transition at $T_c = 1.8$ K well below an antiferromagnetic ordering temperature $T_N = 14.3$ K [1,2]. The U-based HF systems generally involve more than two $5f$ electrons. In UPd$_2$Al$_3$, it is suggested that two of them, that possess a localized character, are responsible for the antiferromagnetism (AFM), whereas the remaining $5f$ electrons, which are hybridized with conduction electrons and become heavy, are responsible for SC. Sato *et al.* have proposed a picture that magnetic excitons resulting from the exchange interactions between magnetic moments produce effective interactions between itinerant electrons and are responsible for the onset of SC [3]. Quite recently, SC was discovered even in the ferromagnetic materials UGe$_2$ and URhGe [4,5]. Although the detailed mechanism for SC has not been identified yet in these materials [6], the fact that SC and ferromagnetism disappear at a same value of critical pressure in UGe$_2$ gives a hint for an intimate relationship between the two types of orderings [4,7].

In contrast to these U-based HF materials, Ce-based HF compounds involve only one $4f$ electron per Ce ion, which may participate in both magnetism and SC. Measurements of resistivity under pressure (P) in CeCu$_2$Ge$_2$, CePd$_2$Si$_2$, and CeIn$_3$ also suggest a coexistence of AFM and SC in a narrow P range [8–10]. It has, however, still remained controversial whether the coexistence of the two phases is homogeneous or they are spatially segregated.

The discovery of a new Ce-based compound CeRhIn$_5$ has opened a way to systematically investigate the evolution from the antiferromagnetic to superconducting state as a function of the pressure [11]. This is because the critical pressure $P_c \sim 1.6$ GPa, at which the two phases meet, is much lower than in the previous examples. Remarkably, its T_c does not only reach a record of $T_c = 2.1$ K to date, but also SC is more robust against increasing pressure than in CePd$_2$Si$_2$ and CeIn$_3$ [11,12]. An effective moment at the paramagnetic state is estimated to be $\mu_{\text{eff}} = 2.38\mu_{\text{B}}$, which is somewhat reduced from a value $2.56\mu_{\text{B}}$ expected for a $4f$ localized model [11]. Below $T_N = 3.8$ K, a nuclear quadrupole resonance (NQR) study indicates the rapid development of an internal field H_{int} induced by the Ce spontaneous moments and a spiral modulation of the Ce moments that is incommensurate with the lattice [13]. A recent neutron diffraction experiment revealed that the reduced Ce magnetic moments of $0.37\mu_{\text{B}}$ at 1.4 K form in a helical spiral structure along the c axis with an incommensurate wave vector $q_M = (1/2, 1/2, 0.297)$ [14]. An extensive NQR study has showed that T_N exhibits a moderate variation, whereas the H_{int} is linearly reduced in $P =$ 0–1.23 GPa, extrapolated to zero at $P^* = 1.6 \pm 0.1$ GPa [15]. Note that this P^* is comparable to $P_c = 1.63$ GPa at which the superconducting signature appears [11]. The NQR experiment indicated that the P induced transition from AFM to SC is of a second-order type, but not of a first-order type that was suggeted by the measurements of resisitivity [11] and specific heat [16]. At $P = 2.1$ GPa apart from P_c or P^*, the nuclear spin-lattice relaxation rate $1/T_1$ reveals a T^3 dependence below T_c, consistent with the existence of the line-node gap [15]. This was also corroborated by the specific heat measurement [16]. In alloying systems such as $\mathrm{CeRh_{1-x}A_xIn_5}$ (A = Ir, Co), the two phases were suggested to coexist in certain concentration ranges of x [17–19]. Apparently, the nature of the P induced transition from AFM to SC has remained controversial in $\mathrm{CeRhIn_5}$, especially near $P_c = 1.63$ GPa where both the phases meet one another.

In this Letter, we report results of ^{115}In-NQR and ac susceptibility (ac-χ) experiments in $\mathrm{CeRhIn_5}$ at $P = 1.75$ GPa close to P_c at zero magnetic field ($H = 0$). We find an onset of the magnetic order at $T_N = 2.5$ K under $P = 1.75$ GPa from a clear split of NQR spectrum due to the spontaneous internal field. Simultaneously, bulk SC below $T_c = 2.0$ K is demonstrated by the observation of the Meissner diamagnetism in ac-χ whose size is the same as in the exclusively superconducting phase in $P = 1.95$–2.15 GPa. We conclude that AFM coexists homogeneously with SC at a microscopic level.

A single crystal of $\mathrm{CeRhIn_5}$ was grown by the self-flux method [11], and was moderately crushed into grains in order to make rf pulses penetrate into samples easily. $\mathrm{CeRhIn_5}$ consists of alternating layers of $\mathrm{CeIn_3}$ and $\mathrm{RhIn_2}$ and hence has two inequivalent In sites per unit cell. The ^{115}In-NQR measurements were made at the In(1) site [13,15,20] which is located on the top and bottom faces of the tetragonal unit cell. The NQR spectrum was obtained by plotting the intensity of spin-echo signal as a function of frequency. The high-frequency ac-χ was carried out by measuring the inductance of an *in situ* NQR coil with the frequency ~2.5 MHz that is not far from the NQR frequency $\nu_Q \sim 7$ MHz. Note that the skin depth for the NQR and ac-χ measurements is comparable to a size of grains (~ 0.1 mm) and the magnetic penetration depth is estimated to be between 5000 to 7000 Å for superconducting $\mathrm{CeTIn_5}$ (T = Ir, Co) [21]. So, the present ac-χ measurement is compatible to the dc-χ one in detecting bulk SC. The hydrostatic pressure was applied by utilizing a BeCu or NiCrAl/BeCu piston-cylinder cell, filled with Daphne oil (7373) as a pressure-transmitting medium. When using liquid as a pressure-transmitting medium to obtain quasihydrostatic pressure, care was taken on some inevitable pressure inhomogeneity in the sample. For our pressure cells, an extent of the spatial distribution in values of pressure $\Delta P/P$ is estimated to be ~3% from a broadening in the linewidth in NQR spectrum, for example, at $P = 1.75$ GPa, $\Delta P \sim 0.05$ GPa.

Figure 1 indicates respective NQR spectra at $T = 2.65$ and 1.45 K at $P = 1.75$ GPa for the $1\nu_Q$ transition which is the lowest one among the four transitions for the In nuclear spin $I = 9/2$. In the paramagnetic state above T_N, only one sharp spectrum is observed as shown in the upper panel. As the temperature decreases, the NQR spectrum starts to broaden below $T_N = 2.5$ K and eventually splits into two peaks as seen in the lower panel. This is interpreted as follows. When the Ce-4f magnetic moments order, they induce spontaneous internal fields H_{int} at the In(1) site, so that the shape of the NQR spectrum is split into two lines. This clear split in the spectrum, which can be fitted by two Lorenztian functions as seen in the lower panel of Fig. 1 (solid curve), persists down to low temperatures below $T_c = 2.0$ K. Note that the separation between the two peaks in the spectrum is directly proportional to the size of magnetically ordered moments [13]. The T dependence of H_{int} at $P = 1.75$ GPa is shown in the inset. A plot of H_{int} against T is fitted to $(1 - T/T_N)^\beta$ with $\beta = 0.5$ (solid line) rather than $\beta = 0.25$ (broken line). The latter behavior was consistent with those at $P = 0$ [13] and at the values of lower pressures [15]. This may suggest that the rapid development of H_{int} becomes slower due to a coexistence between SC and AFM in the vicinity of P_c as demonstrated later. Any single peak that is observed when the system is in the paramagnetic state at $T = 2.65$ K does not remain at the magnetically ordered state at $T = 1.45$ K as shown in Fig. 1. This result gives direct evidence for the onset of homogeneous AFM throughout the whole sample, excluding any possibility of phase segregation at $P = 1.75$ GPa near the border at which the two phases meet. The H_{int} at $P = 1.75$ GPa and $T = 1.45$ K is estimated to be ~80Oe from the separation between the

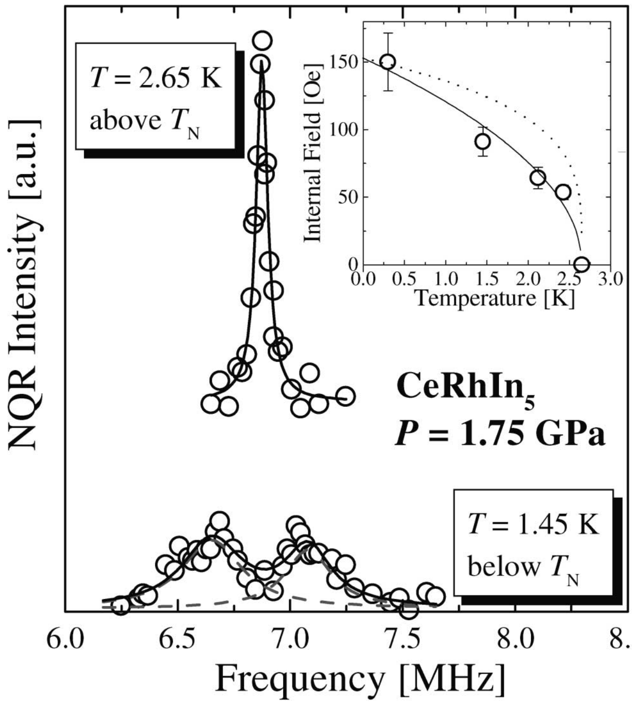

FIG. 1. ^{115}In-NQR spectra of $1\nu_Q$ ($\pm 1/2 - \pm 3/2$ transition for $I = 9/2$) at $P = 1.75$ GPa and $T = 2.65$ K (upper panel) and 1.45 K (lower panel). In the lower panel, the dotted curves are fits by two Lorentzians which peaks are split due to the internal field H_{int} associated with the onset of antiferromagnetism and the solid curve is the convolution of the two curves. The inset shows the temperature dependence of H_{int}. The solid and broken lines are fits to $(1 - T/T_N)^\beta$ with $\beta = 0.5$ and 0.25, respectively.

two peaks, corresponding to a size of the ordered moment whose value is at most ~5% of the value at $P = 0$ [15].

As seen in Fig. 2, the high-frequency ac-χ measurement at $P = 1.75$ GPa has revealed clear bulk SC with its onset temperature at $T_c = 2.0$ K that is lower than $T_N = 2.5$ K. The bulk nature of SC at $P = 1.75$ GPa was warranted by the observation of the same size of the Meissner diamagnetism in ac-χ in $P = 1.95$–2.15 GPa. Note that at $P = 2.1$ GPa unconventional SC with the line-node gap was exclusively established from the measurements of the NQR-T_1 [15] and the specific heat [16]. The data at $P = 1.75$ and 1.95 GPa are actually compared in Fig. 2. By combining this evidence for bulk SC and the emergence of homogeneous internal field due to AFM probed by the NQR spectrum, we conclude that AFM and SC coexist homogeneously at a microscopic level at $P = 1.75$ GPa and hence the P induced transition from AFM to SC around $P_c \sim 1.63$ GPa is not of any first-order type in $CeRhIn_5$, although it was suggested from the measurements of resistivity [11] and specific heat [16].

We summarize the T-P phase diagram for $CeRhIn_5$ in Fig. 3 along with the results reported previously [15,20]. With increasing P, T_N is suppressed at pressures exceeding $P \sim 1$ GPa. Concomitantly the low-energy spectral weight of magnetic fluctuations is suppressed. Namely, the pseudogap behavior emerges as reported in Ref. [20]. Although it is not identified yet whether this behavior is relevant to SC or AFM, the presence of the pseudogap behavior at the border of AFM and SC seems to share a common feature with other strongly correlated electron systems such as high-T_c cuprate and layered organic superconductors [22,23]. The coexistence of AFM and SC in $CeRhIn_5$, however, differs from the previous examples. In particular, the layered organic superconductors show a first-order transition between the two phases [24], as evidenced from the observation of two NMR spectra that arise from the antiferromagnetically ordered phase and superconducting phase near the boundary of the two phases. This coexistence of AFM and SC in $CeRhIn_5$ is unconventional in the context that the HF state derived by one $4f$-electron per Ce ion contributes to both, and hence deserves theoretical studies. A possible scenario for this coexistence may be addressed as follows. When the antiferromagnetic exchange constant $J_{\rm cf}$ between $4f$ electrons and conduction electrons remains strong, $4f$ local moments are compensated due to a Kondo-like interaction and the renormalized HF state could be itinerant, leading to the occurrence of the P induced SC in $CeRhIn_5$. As $J_{\rm cf}$ decreases gradually, the HF quasiparticles tend to polarize since the Ruderman-Kittel-Kasuya-Yosida (RKKY) and the Kondo-like interactions are competing at the quantum critical region where the two phases come across. In such a regime, however, these two effects may lead to the homogeneous coexistence of AFM with tiny ordered moments and SC at a microscopic level.

Finally we note that the interplay between AFM and SC in the first Ce-based compound $CeCu_2Si_2$ that

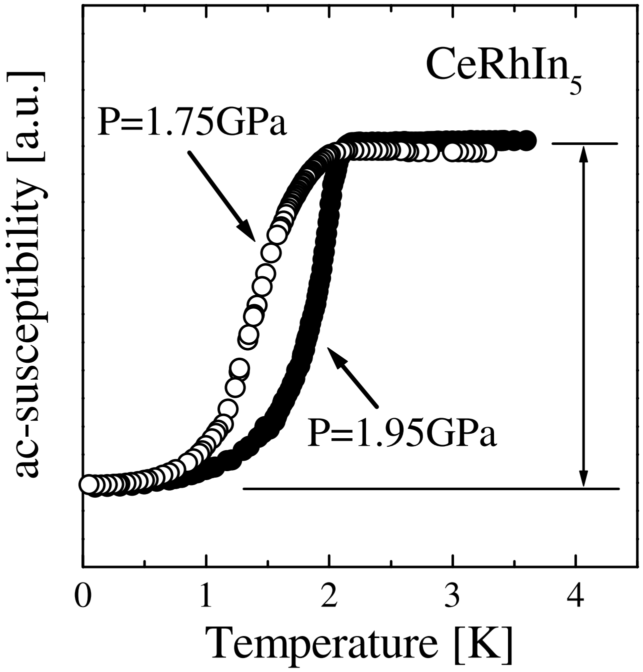

FIG. 2. Temperature dependence of the high-frequency ac susceptibility measured using an *in situ* NQR coil at $P = 1.75$ GPa (open circles) and $P = 1.95$ GPa (closed circles).

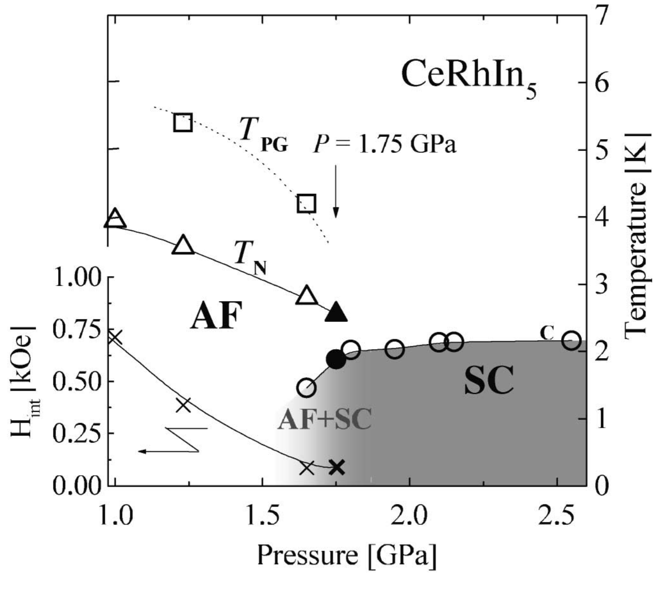

FIG. 3. The temperature-pressure phase diagram for $CeRhIn_5$. The triangles, circles, and crosses show T_N, T_c, and $H_{\rm int}$, respectively, along with the previous results [15,20]. The solid symbols represent the present data ($P = 1.75$ GPa). The open squares, cited from the literature [20], show the T_{PG} at which $1/T_1T$ shows a peak. The solid and broken curves are eye guides. The arrow points the value of pressure.

displays SC at $P = 0$ [25], was recently discussed in the framework of the SO (5) theory [26] that unifies AFM and SC [27] and as a result the two phases are suggested to have a common mechanism. It is worthwhile examining whether the coexistence of the AFM and SC in $CeRhIn_5$ can be accounted for by the SO (5) theory as well.

In conclusion, we have found via the ^{115}In NQR and the ac-susceptibility measurements that AFM and SC coexist homogeneously at a microscopic level in the HF compound $CeRhIn_5$ at $P = 1.75$ GPa. It is unconventional that the Ce $4f$-electron which forms HF state contributes to both AFM and SC. This may be possible in an itinerant regime where the size of ordered moments is largely reduced as actually suggested from the experiment. Our results may also shed light on other strongly correlated systems including high-T_c superconductors.

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