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The phase diagram of antiferromagnetism and superconductivity in $CeRhIn_5$: a study of ^{115}In NQR under pressure

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Abstract
We report on the pressure (P) versus temperature (T) phase diagram of the P-induced heavy fermion (HF) superconductor $CeRhIn_5$ via nuclear quadrupole resonance (NQR) measurements under P. At $P = 2.1$ GPa, the T dependence of the nuclear spin–lattice relaxation rate $1/T_1$ revealed antiferromagnetic critical fluctuations due to the closeness of antiferromagnetism. As P decreases slightly down to 1.9 GPa, a pseudogap emerges above $T_c \sim 2$ K, suggesting the appearance of superconducting fluctuations associated with a strong coupling effect among quasiparticles.

1. Introduction

Due to the competition between Ruderman–Kittel–Kasuya–Yosida interaction and the Kondo effect in heavy fermion (HF) systems, various interesting phenomena emerge near the border of magnetic to non-magnetic phase. In fact, superconductivity (SC) takes place in such a regime for $CeCu_2Si_2$ [1], $CeIrIn_5$ [2, 3] and $CeCoIn_5$ [4, 5]. Pressure (P)-induced SC has been found on the verge of antiferromagnetism (AFM) in $CeCu_2Ge_2$ [6], $CeRh_2Si_2$ [7, 8], $CePd_2Si_2$ [9–11], $CeIn_3$ [9, 10, 12] and $CeRhIn_5$ [13]. An intimate interplay between AFM and SC is evident and even a uniform mixed phase of AFM and SC has been unravelled at their border [14–16].

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$CeRhIn_5$ orders antiferromagnetically at a Néel temperature $T_N = 3.8$ K with an incommensurate wavevector $q_M = (1/2, 1/2, 0.297)$ [17]. A recent neutron experiment revealed the reduced Ce magnetic moments $M_s \sim 0.8\ \mu_B$ [18, 19]. The P-induced transition from AFM to SC takes place at a lower critical pressure $P_c = 1.63$ GPa and higher $T_c = 2.2$ K than in previous examples [6, 7, 9, 11–13].

Previous NQR studies showed that T_N increases gradually up to $P = 1$ GPa, whereas the internal magnetic field (H_{int}) at the In site due to the onset of AFM is linearly reduced in $P = 0$–1.23 GPa, extrapolated to zero at $P^* = 1.6 \pm 0.1$ GPa [20]. Note that this P^* is comparable to $P_c = 1.63$ GPa at which zero resistance was observed [13]. In addition, the temperature (T) dependence of the nuclear spin–lattice relaxation rate ($1/T_1$) shows pseudogap behaviour at $P = 1.23$ and 1.6 GPa [21]. This indicates that $CeRhIn_5$ resembles other strongly correlated electron systems [22, 23]. The T_1 measurement at $P = 2.1$ GPa just above P_c or P^* has probed the development of three-dimensional (3D) antiferromagnetic fluctuations due to the closeness of magnetic criticality, as T cools down to $T_c = 2.2$ K [20]. The observation of $1/T_1 \propto T^3$ below T_c was consistent with the existence of a line-node gap in the SC [20, 24]. This was also corroborated by specific heat measurements under P [25]. The NQR result under P showed that T^*, which is relevant with an energy scale for the hybridization of 4f electrons and conduction electrons [21], gradually increases with a rate of increase of $dT^*/dP \sim 8\text{K GPa}^{-1}$ in $CeRhIn_5$ [21]. The previous report revealed a uniform mixed phase of AFM and SC which was evidenced from the T dependences of the NQR spectrum and ac susceptibility (χ_{ac}) at $P = 1.75$ GPa [15].

In this paper, we report on an interplay between the SC and AFM in $CeRhIn_5$ through ^{115}In NQR experiments at $P = 1.6$, 1.9, 2.0 and 2.1 GPa. Bulk superconductivity which has the mean-field (MF) type of gap growth sets in below $T_c^{MF} = 0.9$ and 2.0 K at $P = 1.6$ and 1.9 GPa, respectively [16]. Furthermore, the T dependence of $1/T_1$ has probed the critical slowing down of magnetic correlations associated with the onset of long range AFM. These results have revealed that the uniform mixed phase of AFM and SC exists in $P = 1.53$–1.9 GPa. We highlight that the magnetic and superconducting characteristics at $P = 2.0$ GPa are unconventional, exhibiting a crossover from the SC with a line-node gap affected by antiferromagnetic critical fluctuations at $P = 2.1$ GPa to the exotic mixed phase of SC and AFM with gapless magnetic and quasi-particle excitations affected by the emergence of pseudogap behaviour above T_N in $P = 1.53$–1.9 GPa.

2. Experimental details

A single crystal of $CeRhIn_5$ was grown by the self-flux method, and was moderately crushed into grains in order to make rf pulses penetrate into samples easily. $CeRhIn_5$ consists of alternating layers of $CeIn_3$ and $RhIn_2$ and hence has two inequivalent In sites per unit cell. The ^{115}In NQR measurements were made at the In(1) site [17, 20, 21] which is located on the top and bottom faces of the tetragonal unit cell. The NQR spectrum was obtained by plotting the intensity of the spin-echo signal as a function of frequency. Measurement of $1/T_1$ was made by the conventional saturation–recovery method in the T ranges of 0.05–100 K and 1.4–100 K at $P = 1.6$ GPa and $P = 1.9$ and 2.0 GPa, respectively. The ^{115}In NQR $1/T_1$ was measured at the transition of $2\nu_Q$ ($\pm 3/2 \leftrightarrow \pm 5/2$) above $T = 1.4$ K, but at $1\nu_Q$ ($\pm 1/2 \leftrightarrow \pm 3/2$) below $T = 1.4$ K. The hydrostatic pressure was applied by utilizing a BeCu piston–cylinder cell, filled with Daphne oil (7373) as a pressure-transmitting medium. When using liquid as a pressure-transmitting medium to obtain quasi-hydrostatic pressure, care was taken with some inevitable pressure inhomogeneity in the sample. For our pressure cells, the extent of the spatial distribution in values of pressure $\Delta P/P$ is estimated to be $\sim$3% over the whole sample from the broadening in the linewidth in the NQR spectrum.

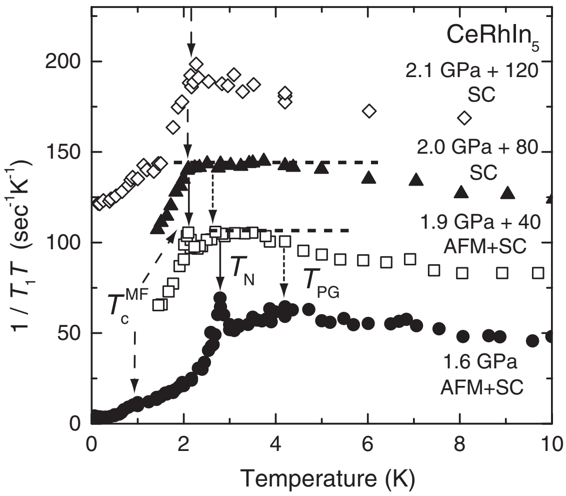

Figure 1. The offset T dependence of $1/T_1$ at $P = 1.6$, 1.9, 2.0 and 2.1 GPa. Dashed, solid and dotted arrows indicate T_{PG}, T_N and T_c^{MF}, respectively. Dotted lines indicate the relation $1/T_1T$ = constant. Note that the crossover behaviour, from the pseudogap (AFM) to the nearly 3D antiferromagnetic spin fluctuation regime (SC), is observed between $P = 1.9$ and 2.1 GPa (see the text).

3. Experimental result

Figure 1 indicates the T dependence of $1/T_1T$ at $P = 1.6$, 1.9, 2.0 and 2.1 GPa. Here the respective values of $1/T_1T$ are scaled up with the offset values of 40, 80 and 120 s^{-1} K^{-1} at $P = 1.6$, 1.9, 2.0 and 2.1 GPa. At pressures exceeding $P = 1.53$ GPa, the onset of bulk SC was confirmed from the measurements $1/T_1T$ and χ_{ac} as reported previously [16]. Pseudogap behaviour is suggested by a tiny peak in $1/T_1T$ at $T_{PG} = 4.2$ and 2.7 K at $P = 1.6$ and 1.9 GPa, respectively. The faint $1/T_1T$ = constant behaviour below 4 K at $P = 2.0$ GPa is associated with a coincidental combined effect of the decrease in $1/T_1T$ due to the pseudogap and the increase due to the development of antiferromagnetic fluctuations. In fact, the $1/T_1T$ at $P = 2.1$ GPa continues to increase down to $T_c^{MF} = 2.2$ K [20]. One scenario for explaining this crossover behaviour in magnetic excitations at $P = 2.0$ GPa is that the strong coupling effect for superconducting pairing formation tends to suppress low lying magnetic excitations as the system approaches antiferromagnetic criticality.

What happens when entering the antiferromagnetic regime as P decreases slightly from 2.0 to 1.9 GPa? At $P = 1.9$ GPa, the SC sets in at $T_c^{MF} = 2.0$ K [16], and the onset of AFM is evidenced by a clear peak in $1/T_1T$ at $T = 2.0$ K due to the critical slowing down of magnetic correlations as well as being seen at $T_N = 2.8$ K at $P = 1.6$ GPa. Therefore, the AFM emerges suddenly below $T_N = 2.0$ K at $P = 1.9$ GPa, notably coinciding with the onset of SC. Unfortunately, however, the appearance of a tiny value of H_{int} at the In(1) site and the superconducting diamagnetism prevent us from detecting NQR signals below $T = 1$ K at all. So, it remains as future work to establish what kinds of superconducting and magnetic properties are realized in such a uniform mixed phase of AFM and SC below $T_N = T_c = 2.0$ K at $P = 1.9$ GPa. More systematic experiments between $P = 1.9$ and 2.1 GPa are required to unravel the intimate interplay of AFM and SC in $CeRhIn_5$.

Figure 2 shows the novel phase diagram of AFM and SC in $CeRhIn_5$ determined from the NQR experiments under P. As shown shaded in figure 2, the uniform mixed phase of AFM

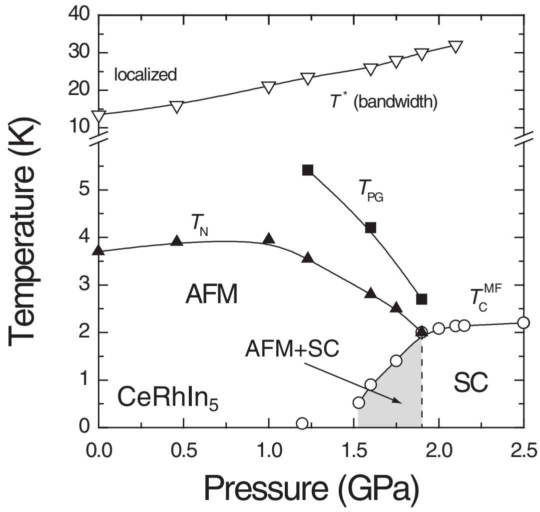

Figure 2. The novel P–T phase diagram of $CeRhIn_5$. The shaded region indicates the microscopic coexistence of AFM and SC. The dotted line at $P = 1.9$ GPa suggests the phase boundary of AFM at zero magnetic field (see the text).

and SC is established in $P = 1.53$–1.9 GPa from the extensive measurements of $1/T_1$, the NQR spectrum and χ_{ac} [16]. At $P = 1.9$ GPa, the AFM and SC merge at $T_N = T_c = 2.0$ K. The uniform mixed phase of AFM and SC that accompanies the pseudogap crosses over to the SC regime with the line-node gap affected by strong antiferromagnetic fluctuations around $P = 2.0$ GPa.

4. Summary

We have presented the P–T phase diagram of the HF antiferromagnet $CeRhIn_5$ via ^{115}In NQR measurements. The uniform mixed phase of AFM and SC has been established in $P = 1.53$–1.9 GPa. It has been demonstrated that the AFM and SC merge at $T_N = T_c = 2.0$ K at $P = 1.9$ GPa. SC with a line-node gap arises at $P = 2.1$ GPa, exhibiting strong antiferromagnetic fluctuations. Therefore, the uniform mixed phase of AFM and SC crosses over to a single phase of SC in a very narrow P range, 1.9–2.1 GPa. Here we have presented a magnetic critical point located around $P = 2.0$ GPa where the onset of AFM is not evident at zero magnetic field. Further experiments are required to understand these novel phases of matter on the basis of the magnetic criticality in $CeRhIn_5$.

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