Pressure-Temperature Phase Diagram in Ce$_2$RhIn$_8$
Studied by In-NQR Measurements

M. Yashima$^{1,2}$, S. Taniguchi$^1$, H. Miyazaki$^1$, H. Mukuda$^{1,2}$, Y. Kitaoka$^1$, H. Shishido$^{3,5}$, R. Settai$^3$, and Y. Ônuki$^{3,4}$

$^1$Department of Materials Engineering Science, Osaka University, Osaka 560-8531, Japan
$^2$JST, TRIP (Transformative Research-Project on Iron Pnictides), Chiyoda, Tokyo 102-0075, Japan
$^3$Department of Physics, Osaka University, Osaka 560-0043, Japan
$^4$Advanced Science Research Center, Japan Atomic Energy Research Institute, Tokai, Ibaraki 319-1195, Japan
E-mail: mitsuharu@nmr.mp.es.osaka-u.ac.jp

Abstract. We present a phase diagram for the antiferromagnetism and superconductivity in Ce$_2$RhIn$_8$ probed by In-NQR studies under pressure ($P$). The quasi-2D character of antiferromagnetic spin fluctuations in the paramagnetic state at $P = 0$ evolves into a 3D character because of the suppression of antiferromagnetic order for $P > P_{QCP} \sim 1.36$ GPa (QCP: antiferromagnetic quantum critical point). Nuclear-spin-lattice-relaxation rate $1/T_1$ measurements revealed that the superconducting order occurs in the $P$ range 1.36 – 1.84 GPa, with maximum $T_c \sim 0.9$ K ($P_{QCP} \sim 1.36$ GPa).

1. Introduction
The heavy-fermion (HF) compounds CeIn$_3$ [1] and CeTIn$_5$ (T = Co, Rh) [2, 3, 4] revealed an intimate relationship between antiferromagnetism (AFM) and superconductivity (SC). CeIn$_3$ has a cubic crystal structure, and it is expected to exhibit the three-dimensional (3D) magnetic interaction. CeIn$_3$ is an antiferromagnet with $T_N = 10$ K at ambient pressure ($P = 0$), and AFM discontinuously collapses around $P_c = 2.46$ GPa, suggesting that the quantum phase transition from AFM to paramagnetism (PM) is of the first order [5, 6]. It was suggested that the first-order quantum phase transition is responsible for the occurrence of SC in CeIn$_3$ [6]. CeRhIn$_5$, which has a tetragonal crystal structure, is also an antiferromagnet with $T_N = 3.8$ K at $P = 0$ [3]. For CeRhIn$_5$, we have shown that the tetracritical point, where the AFM, AFM+SC, SC, and PM phases are in contact, exists at $P_{tetra} \sim 1.98$ GPa and $T_c$ reaches the maximum value ($\sim 2.2$ K) at approximately 2.5 GPa from the AFM quantum critical point (QCP), which lies at $P_{QCP} \sim 2.1$ GPa [7]. CeTIn$_5$, Ce$_2$TIn$_8$ and, CeIn$_3$ (T = Co, Rh, Ir) are a series of structurally related materials with chemical compositions of the form Ce$_m$TIn$_{3m+2}$ with $m = 1, 2, \infty$, respectively. Ce$_2$TIn$_8$ enables us to study the relationship between the structure-based evolution of magnetic characteristics and the onset of unconventional SC in HF systems.

Ce$_2$RhIn$_8$ is an antiferromagnet with $T_N = 2.8$ K at $P = 0$ and a magnetic moment of 0.55 $\mu_B$ per Ce ion [8]. The pressure-temperature ($P-T$) phase diagrams of Ce$_2$RhIn$_8$ reported thus
far are based on resistivity, ac-susceptibility, and heat-capacity measurements [9, 10, 11, 12]. The resistivity measurements revealed that as $P$ increases, $T_N$ monotonously decreases down to 1.2 K at 1.5 GPa; further, SC occurs for $P > 1$ GPa and exhibits the maximum $T_c$ ($T_c^{\text{max}} \sim 2$ K) around 2.3 GPa. On the other hand, the heat-capacity measurements indicated that an AFM order survives up to $P = 1.65$ GPa, but no anomalies that signal the onset of SC were observed. The previously reported NQR-$1/T_1$ measurement was performed to investigate the onset of SC with $T_c = 0.9$ K at $P = 1.87$ GPa [13]. In this context, a $P - T$ phase diagram for Ce$_2$RhIn$_8$ is not yet fully understood.

2. Experimental Procedure

For obtaining NQR measurements, Ce$_2$RhIn$_8$ grown by the self-flux method was moderately crushed into a coarse powder to allow RF pulses to easily penetrate the sample. Hydrostatic pressure was applied using a NiCrAl-BeCu piston-cylinder cell filled with a Si-based organic liquid as the pressure-transmitting medium. To calibrate the pressure at low temperatures, the shift in the $T_c$ of Sn metal was monitored by using the resistivity measurements. The crystal structure of Ce$_2$RhIn$_8$ consists of alternating layers of CeRhIn$_5$ and CeIn$_3$. There are three In sites per unit cell, denoted by In(1), In(2), and In(3). In(1) and In(2) are located in the CeRhIn$_5$ layer and In(3) is located in the CeIn$_3$ layer. In(2) is surrounded by two Ce and two Rh ions. The measurements for the $^{115}$In-NQR ($I = 9/2$) spectrum were mainly performed at the $3\nu_Q$ transition at In(2) in Ce$_2$RhIn$_8$. Here, $\nu_Q$ is defined by the NQR Hamiltonian, $\mathcal{H}_Q = (h\nu_Q/6)[3I_z^2 - I(I + 1) + \eta(I_x^2 - I_y^2)]$, where $\eta$ is the asymmetry parameter of the electric field gradient. Using $\nu_Q = 16.41$ MHz and $\eta = 0.43$, the NQR frequency of the $3\nu_Q$ transition is estimated as 47.4 MHz for In(2) at $P = 0$.

3. Results and discussion

Fig. 1a shows the $T$ dependence of $1/T_1$ at high $T$ and $P = 0 - 2.27$ GPa in Ce$_2$RhIn$_8$. A distinct peak in $1/T_1$ is associated with the onset of AFM order at $T_N = 2.85$ K and $P = 0$ GPa. Note that in the PM state, $1/T_1$ increases up to 200 K at $P = 0$, suggesting that Ce-derived magnetic fluctuations occur in an itinerant regime; this is consistent with the NQR measurement results [13] and the angle-resolved photoemission spectroscopy results [14]. The behavior $1/T_1 \propto T^{1/4}$ is consistent with a quasi-2D AFM spin-fluctuations (SFs) model that predicts the relation $1/T_1T \propto \chi_Q(T)^{3/4}$ near an AFM QCP. This $T^{1/4}$-behavior of $1/T_1$ resembles that at ambient $P$ in CeCoIn$_5$ [15]. Here, the term quasi-2D AFM SFs implies that the magnetic correlation length in the tetragonal plane develops at a faster rate than that along the c-axis and that the staggered susceptibility $\chi_Q(T)$ with the AFM wave number $Q = (\pi, \pi, \pi)$ is anticipated to obey the Curie-Weiss law as $\chi_Q(T) \propto 1/(T + \theta)$. In this context, it is predicted that the quasi-2D AFM SFs will obey $1/T_1 \propto T \times \chi_Q(T)^{3/4} \propto T^{1/4}$ in the vicinity of the AFM QCP, where $\theta \sim 0$. As $P$ increases, the $T_N$ determined from a peak in $1/T_1$ decreases to $T_N = 1.2$ K at $P = 0.92$ GPa. At $P = 1.36$ GPa, a marked decrease in $1/T_1$ below 0.9 K without an accompanying peak was observed. Since the broadening of the NQR spectrum due to the internal field was not observed (not shown here), the AFM order disappears at $P_{\text{QCP}} \sim 1.36$ GPa. Furthermore, the SC diamagnetism was observed above 1.36 GPa from the ac-susceptibility measurements (not shown here). In this context, the anomaly in $1/T_1$ at 1.36 GPa suggests that SC sets in below $T_c = 0.9$ K.

Note that as $P$ increases, the behavior $1/T_1 \propto T^{1/4}$ at $P = 0$ evolves into $1/T_1 \propto T^{1/2}$ above $P_{\text{QCP}}$, as shown in Fig. 1a. The latter relation is consistent with the 3D-AFM SFs model that predicts the relation $1/T_1T \propto \chi_Q(T)^{1/2}$ near the 3D-AFM QCP. This $P$-evolution of $1/T_1$ indicates that the crossover from the quasi-2D to 3D character of AFM SFs occurs between $P = 0$ and 1.36 GPa.
Figure 1. $T$ dependences of $1/T_1$ at (a) high $T$ and (b) at low $T$ for $P = 0 - 2.27$ GPa in Ce$_2$RhIn$_8$. Solid and dashed arrows point to $T_N$ and $T_c$, respectively. The inset shows the $T$ dependence of ac-susceptibility at $P = 1.36, 1.62, 1.84$, and $2.27$ GPa in the order indicated by the direction of the arrow.

Figure 2. The $P - T$ phase diagram of Ce$_2$RhIn$_8$ obtained from the NQR measurements.

It is remarkable that SC with $T_{\text{max}} = 0.9$ K are observed at $P_{\text{QCP}} = 1.36$ GPa, as shown in Fig. 2. These results suggest the intimate relationship between the unconventional SC and the AFM QCP in Ce$_2$RhIn$_8$. Furthermore, it should be noted that SC sets in as a result of the evolution from the quasi-2D to 3D character of AFM SFs. This is in contrast to the fact that the SC dome in the Ce115 compounds with $T_{\text{c max}} > 2$ K is realized around the quasi-2D AFM QCP but is separated from the phase boundary between the AFM and PM phases. These results demonstrate the intimate relationship between the dimensionality of AFM SFs and the onset of unconventional SC; the 2D character of AFM SFs is favorable to the increase of the $T_c$ in HF SC compounds as well as in high-$T_c$ copper oxides.

In conclusion, we have established the $P - T$ phase diagram for Ce$_2$RhIn$_8$ from microscopic In-NQR measurements. The AFM order disappears at $P_{\text{QCP}} \sim 1.36$ GPa, where 3D-AFM SFs are dominant. It was demonstrated that the SC order occurs in the narrow $P$ range of 1.36 – 1.84.
GPa and exhibits $T_{\text{max}} = 0.9$ K around $P_{QCP} \sim 1.36$ GPa. We state that this phase diagram differs from the previously reported ones because the latter were affected by contamination by impurity phases such as CeRhIn$_5$. The unconventional SC in Ce$_2$RhIn$_8$ occurs under the development of 3D AFM SFs rather than the quasi-2D AFM SFs, as in the case of CeCoIn$_5$ and CeRhIn$_5$. Noting that the $T_c = 0.9$ K for Ce$_2$RhIn$_8$ is significantly lower than the $T_c (> 2$ K) for CeCoIn$_5$ and CeRhIn$_5$, it is suggested that the 2D character of AFM SFs plays a vital role in increasing the $T_c$ in strongly correlated electron systems.

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