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Pressure-temperature phase diagram of antiferromagnetism and superconductivity in $CeRhIn_5$ and $CeIn_3$: ^{115}In-NQR study under pressure

S. Kawasaki,[1] T. Mito,[1] G.-q. Zheng,[1] C. Thessieu,[1,*] Y. Kawasaki,[1] K. Ishida,[1] Y. Kitaoka,[1] T. Muramatsu,[1] T. C. Kobayashi,[2] D. Aoki,[3] S. Araki,[3] Y. Haga,[4] R. Settai[3] and Y. Onuki[3,4]

[1]*Department of Physical Science, Graduate School of Engineering Science, Osaka University, Osaka 560-8531, Japan*

[2]*Research Center for Materials Science at Extreme Conditions, Osaka University, Toyonaka, Osaka 560-8531, Japan*

[3]*Department of Physics, Graduate School of Science, Osaka University, Toyonaka, Osaka 560-0043, Japan*

[4]*Advanced Science Research Center, Japan Atomic Energy Research Institute, Tokai, Ibaraki 319-1195, Japan*

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We report the pressure (P)-temperature (T) phase diagram of antiferromagnetism and superconductivity in $CeRhIn_5$ and $CeIn_3$ revealed by the ^{115}In nuclear-spin-lattice-relaxation (T_1) measurement. In the itinerant magnet $CeRhIn_5$, we found that the Néel temperature T_N is reduced at $P \geqslant 1.23$ GPa with an emergent pseudogap behavior. In $CeIn_3$, the localized magnetic character is robust against the application of pressure up to $P \sim 1.9$ GPa, beyond which the system evolves into an itinerant regime in which the resistive superconducting phase emerges. We discuss the relationship between the phase diagram and the magnetic fluctuations.

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It has been reported that a superconducting (SC) order in cerium (Ce)-based heavy-fermion (HF) compounds takes place nearby the border at which an antiferromagnetic (AF) order is suppressed by applying pressure (P) to the HF-AF compounds $CeCu_2Ge_2$,[1] $CePd_2Si_2$ (Ref. 2) and $CeIn_3$.[3] The superconductivity in these compounds, however, occurs only in extreme conditions where the pressure exceeds ~ 2 GPa and temperature (T) is cooled down below ~ 1 K. Indeed the experiments were restricted mainly to transport measurements. The discovery of P-induced HF superconductors in Ce-based HF-AF compounds has stimulated further experimental works under P.[4–7] In order to gain profound insight into a relationship between magnetism and superconductivity in HF systems, systematic NMR/NQR experiments under P are important, since they can probe the evolution of the magnetic properties toward the onset of SC phase.

Recently, Hegger *et al.* found that a new HF material $CeRhIn_5$ consisting of alternating layers of $CeIn_3$ and $RhIn_2$ reveals an AF-to-SC transition at a relatively lower critical pressure $P_c = 1.63$ GPa than in all previous examples.[1–3] The SC transition temperature $T_c = 2.2$ K is the highest one to date among P-induced superconductors.[4] This finding has opened a way to investigate the P-induced evolution of both magnetic and SC properties over a wide P range. In a previous paper,[7] the ^{115}In NQR study of $CeRhIn_5$ has clarified the P-induced anomalous magnetism and unconventional superconductivity. In the AF region, the Néel temperature T_N exhibits a moderate variation, while the internal field H_{int} at the ^{115}In(1) site in the $CeIn_3$ plane due to the magnetic ordering is linearly reduced in $P = 0$–1.23 GPa, extrapolated to zero at $P^* = 1.6 \pm 0.1$ GPa. This P^* is comparable to $P_c = 1.63$ GPa at which the SC signature appears,[4] which was indicative of a second-order-like AF-to-SC transition rather than the first-order one suggested previously.[4] At $P = 2.1$ GPa, it was found that the nuclear spin-lattice-relaxation rate $1/T_1$ reveals a T^3 dependence below the SC transition temperature T_c, which shows the existence of line-nodes in the gap function.[7] It is, however, not yet clear how the electronic states change with P when the AF phase evolves into the SC phase.

On the other hand, $CeIn_3$ crystallizes in the cubic $AuCu_3$ structure and orders antiferromagnetically ($T_N = 10$ K) at $P = 0$ with an ordering vector $\mathbf{Q} = (1/2,1/2,1/2)$.[8] T_N monotonically decreases with P and superconductivity appears below $T_c \sim 0.2$ K at a critical pressure $P_c = 2.55$ GPa, but the onset of the superconductivity was observed *only* by the resistivity measurement and is limited in a narrow P range of about 0.5 GPa.[2,3,9] The previous ^{115}In-NQR result on $CeIn_3$ revealed that the AF order disappears around $P^* = 2.44$ GPa close to P_c and the Fermi-liquid behavior was observed below 10 K at $P = 2.74$ GPa.[10] However, no SC transition was seen. Thus, the bulk nature of the P-induced SC state in $CeIn_3$ has not been confirmed yet by other measurements than resistivity. This is in contrast with the case of $CeRhIn_5$ where the bulk nature of the SC phase was fully established[7] which continues up to 8 GPa.[11]

Here we report P-T phase diagrams for $CeRhIn_5$ and $CeIn_3$ obtained through ^{115}In-T_1 measurements and present a possible explanation for the different P dependence of the SC phase in the two compounds.

The single crystals of $CeRhIn_5$ and $CeIn_3$ were grown by the self-flux and Czochralski Method, respectively,[4,12] and were moderately crushed into grains in order to make rf pulses penetrate into samples easily. In a small piece of $CeIn_3$, the zero resistance transition is confirmed in $P = 2.2$–2.8 GPa (Ref. 11) (see below for detail). At $P = 2.45$ GPa, T_c reaches a maximum of $T_c = 0.19$ K and its transition width is the sharpest, which is in accordance with the previous results.[2,3] The T_1 was measured at the transitions of $2\nu_Q(\pm 3/2 \leftrightarrow \pm 5/2)$ and $3\nu_Q(\pm 5/2 \leftrightarrow \pm 7/2)$ for the ^{115}In(1) site in $CeRhIn_5$ and of $3\nu_Q$ in $CeIn_3$, by the conventional saturation-recovery method. The hydrostatic pressure was applied by utilizing a BeCu piston-cylinder cell, filled with Daphne oil (7373) as a pressure-transmitting medium.

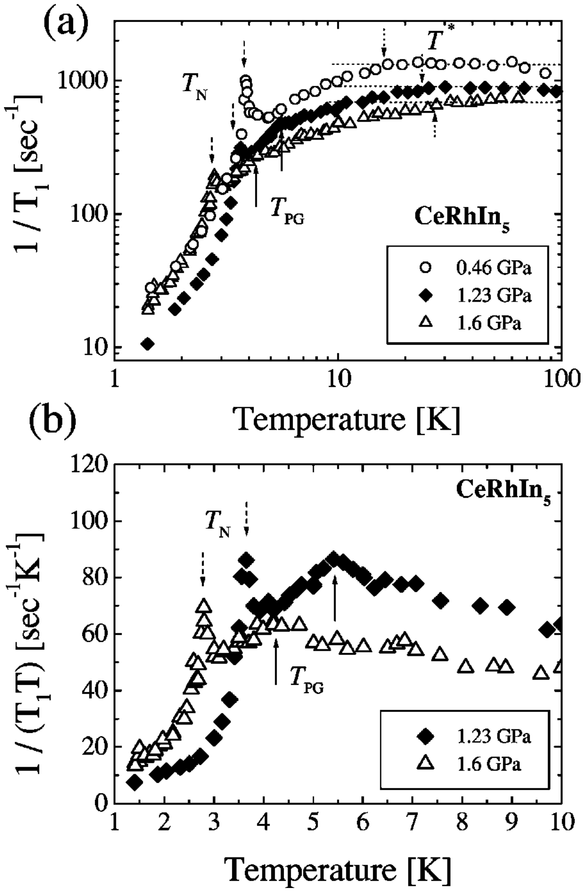

FIG. 1. (a) T dependence of $^{115}(1/T_1)$ in $CeRhIn_5$ at $P=0.46$, 1.23, and 1.6 GPa. The dotted lines are eye-guides. The dotted arrow indicates T^*. (b) T dependence of $1/T_1T$. In both (a) and (b), the solid and broken arrows indicate T_{PG} and T_N, respectively.

Figure 1(a) shows some typical data sets for the T dependence of $1/T_1$ in $CeRhIn_5$ at $P=0.46$, 1.23 and 1.6 GPa. In the high-T region, it is notable that $1/T_1$ becomes almost T independent above the temperature marked as T^*. This indicates that Ce-4f moment fluctuations are in a localized regime above T^*. In such a regime, $1/T_1$ is proportional to $p_{\rm eff}^2/J_{\rm ex}$ or $\sim p_{\rm eff}^2 W/J_{\rm cf}^2$ in terms of a localized moment picture. Here $p_{\rm eff}$ is an effective paramagnetic local moment, $J_{\rm ex}$ the Ruderman-Kittel-Kasuya-Yosida exchange constant among 4f moments, $J_{\rm cf}$ the exchange constant between 4f moments and conduction-electron spin, and W is the bandwidth (BW) of conduction electrons. A progressive suppression in $1/T_1$ with increasing P is considered as due to the reduction in $p_{\rm eff}$ and/or the enhancement of $J_{\rm cf}$. It is known in HF systems that T^* is scaled to the quasielastic linewidth in the neutron-scattering spectrum, leading to a tentative estimation of the BW of the HF state. In $CeRhIn_5$, therefore, the incommensurate spiral order with a staggered moment of $0.267\mu_B$ occurs in an itinerant magnetic regime at $P=0$.[15,17] As seen in Fig. 1(a), T^* increases progressively with $T^*=16$, 23, and 27 K at $P=0.46$, 1.23, and 1.6 GPa, respectively. Since the application of P further increases the hybridization between Ce-4f state and conduction electrons, $J_{\rm cf}$, $J_{\rm ex}$, T^*, and BW are increased, whereas the size of the ordered moments is reduced. In this case, one could expect that the slight increase in $T_N(P)\propto p_{\rm eff}^2(P)J_{\rm ex}(P)$ with increasing pressure is due to the enhancement of $J_{\rm ex}(P)$ which overcomes a possible reduction in $p_{\rm eff}$.

In order to focus on the itinerant magnetic regime in $T<T^*$, the T dependence of $1/T_1T$ below 10 K is shown in Fig. 1(b) for $P=1.23$ and 1.6 GPa. Above T_N, it is clearly seen that $1/T_1T$ shows a broad peak at T_{PG}. This resembles the pseudogap behavior found in the high-T_c copper oxide superconductors.[16] Namely, when P approaches the critical pressures P^* or P_c, the low-energy spectral weight of magnetic fluctuations is suppressed before an ordering occurs. This is an observation of the pseudogap behavior in a magnetic region close to the SC phase. We note that the pseudogap behavior has been found in either two- or lower-dimensional strongly correlated electron systems.[16] Very recently, anisotropic 3D AF fluctuation was reported from neutron scattering with an energy scale of less than 1.7 meV for temperature as high as 3 T_c.[17] In fact, an anisotropic 3D character in the AF fluctuations has been revealed previously by NQR measurement in the related material $CeIrIn_5$ that is a superconductor at $P=0$.[18] Thus the observation of the pseudogap behavior is consistent with the system bearing a magnetic character of reduced dimensionality at low pressures. On the other hand, when the pressure is further increased, at $P=2.1$ GPa where the bulk SC transition appears, for example, $1/T_1T$ continues to increase down to $T_c=2.2$ K without any signature for the pseudogap behavior.[7] The T variation of $1/T_1T$ is consistent with the three-dimensional (3D) AF Fermi-liquid model of the self-consistent renormalized (SCR) theory for nearly AF metals.[13] Thus an evolution in the magnetic fluctuations, from a magnetic regime of reduced dimensionality to a more isotropic one, may take place in a narrow P window of 1.2–2.1 GPa when the magnetic order evolves into the SC order.

Next we deal with the results in $CeIn_3$. Figure 2(a) represents the T dependence of $1/T_1$ in $CeIn_3$ at $P=0$, 1.42, 2.35, and 2.65 GPa. It is seen clearly that the T and P dependencies of $1/T_1$ differ from $CeRhIn_5$. $1/T_1$ above T_N is nearly T independent at $P=0$. At $P=1.42$ GPa, $1/T_1$ even increases upon cooling. Thus, the localized magnetic character in the paramagnetic state is robust against the application of P in $CeIn_3$. In the high pressure regime where the SC sets in, there seems to appear an itinerant regime below $T^*\sim 10$ K. T^* eventually reaches ~ 30 K at $P=2.65$ GPa. Figure 2(b) indicates the T dependence of $1/T_1T$. For all pressures, $1/T_1T$ increases down to T_N, giving no indication for the pseudogap behavior. At $P=2.35$ GPa where the zero resistance is observed, it has been confirmed that the magnetically ordering survives with a relatively high value of $T_N=5$ K. The SC transition is, however, not found by the high-frequency ac susceptibility measurement down to 100 mK using the *in situ* NQR coil, despite that a zero resistance was observed in the same sample at $T_c\sim 0.15$ K. From the present experiments, we conclude that the P-T phase diagram and the nature of the SC phase in $CeIn_3$ differs from in $CeRhIn_5$, in many aspects, reflecting their contrasting electronic and magnetic properties; we speculate that the SC

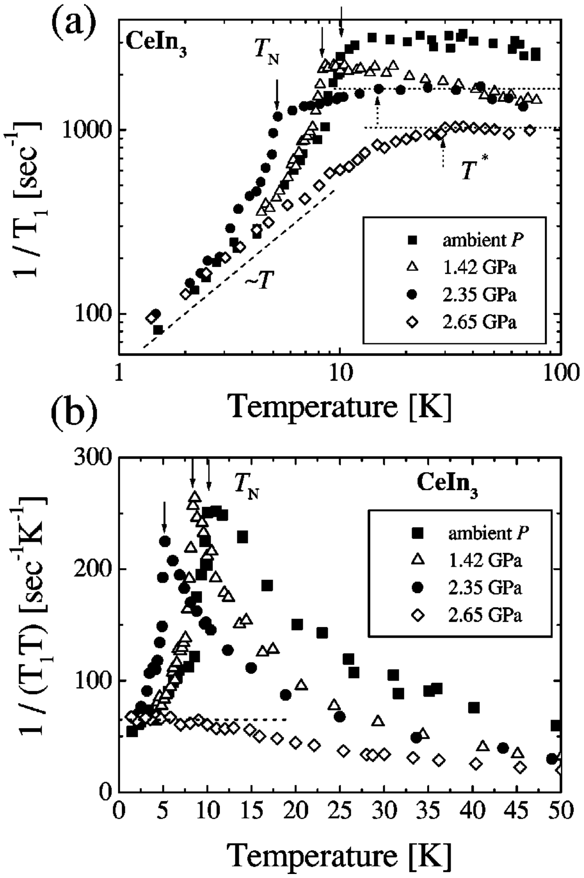

FIG. 2. (a) T dependence of $^{115}(1/T_1)$ in $CeIn_3$ at $P=0$, 1.42, 2.35, and 2.65 GPa. The dotted and broken lines are eye-guides. The dotted arrow indicates T^*. (b) The T dependence of $1/T_1T$. In both (a) and (b), the solid arrows indicate T_N.

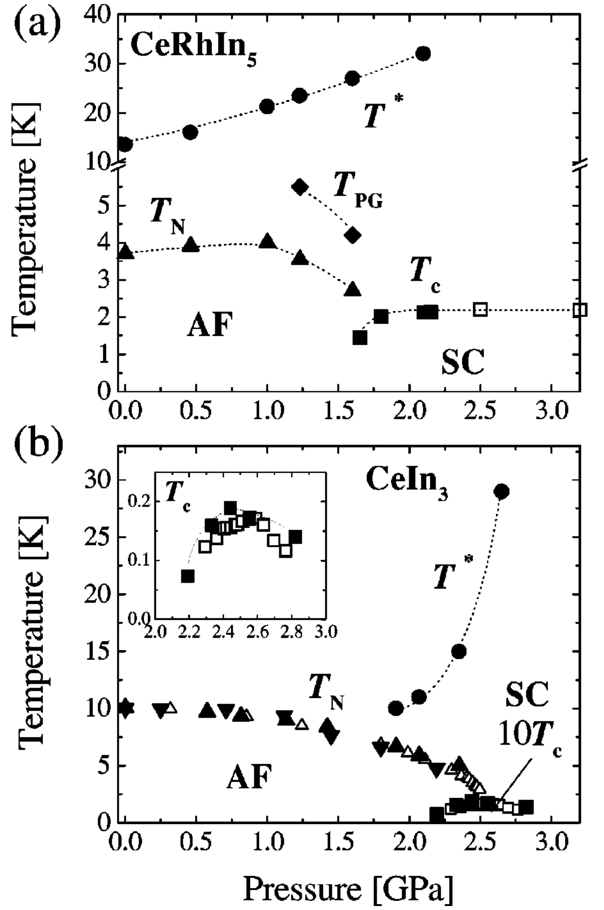

FIG. 3. P-T phase diagrams (a) for $CeRhIn_5$ and (b) for $CeIn_3$. (a) The open marks are determined from the resistivity measurements (Ref. 11), and the solid squares are determined from the ac-χ measurement.(Ref. 7) (b) The open marks for T_N and T_c are taken from the resistivity measurements (Refs. 2 and 3). The rest marks are determined from the present work. The inset indicates the detailed P dependence of T_c in expanded scales.

phase in $CeIn_3$ that accompanies the Meissner diamagnetism, if any, is even narrower than suggested by the resistivity measurement.

The P-T phase diagrams for $CeRhIn_5$ and $CeIn_3$ are summarized in Fig. 3. For $CeRhIn_5$, T^* slightly increases with increasing P up to 1.0 GPa, as does T_N which coincides with the previous result.[4] However, T_N decreases above $P = 1.23$ GPa. At the same time, a pseudogap behavior emerges below $T_{PG} \sim 5.5$ K. As P is further increased, T^* moderately increases with $dT^*/dP \sim 8$ K/GPa. It is noted that T^* is comparale to the temperature $T_{max,\rho}$ at which the resistivity exhibits a maximum value and also $dT^*/dP \sim dT_{max,\rho}/dP$.[4] We remark that T^* and dT^*/dP are also close to those in $CeCu_2Si_2$ (Ref. 6) which is a superconductor at $P=0$ and reveals a SC state over a wide P region as well.[19] In $CeRhIn_5$, the anisotropic 3D AF fluctuations may survive until the system is close to the SC state, where the pseudogap behavior is emergent. Above P^* or P_c where the bulk superconductivity appears, the AF spin correlations become more isotropic and the T dependence of $1/T_1T$ for $T > T_c$ can be explained on the basis of the 3D SCR theory for nearly AF metals.[7] While the isotropic AF fluctuation regime is fully established, the bulk SC is insensitive against P. In $CeIn_3$, by contrary, T^* steeply increases above $P = 1.9$ GPa, indicating an evolution of the system into an itinerant magnetic regime. It is interesting that the SC state becomes emergent in such a regime. Close to P_c, the normal state resistivity, ρ in $CeIn_3$ at low T is also consistent with the 3D AF fluctuations model of the itinerant magnetic regime.[3] However, when the pressure is further increased above P_c, a Fermi-liquid behavior of the resistivity returns more rapidly in $CeIn_3$ than in $CeRhIn_5$ and $CeCu_2Si_2$.[3] This is also corroborated by the observation of T_1T = constant behavior at $P = 2.65$ GPa in $CeIn_3$. Thus the window of the 3D AF fluctuation regime is much narrower in $CeIn_3$. We propose that the narrow SC region in $CeIn_3$ is due to the small window for the 3D AF fluctuation regime because of its large rate of dT^*/dP. This small window for the 3D AF fluctuation regime against P in $CeIn_3$ may be related to its cubic crystal structure which is more sensitive to external P than a tetragonal structure.

Based on magnetically mediated SC theoretical models,[20] it is predicted that 2D AF fluctuations are superior to 3D fluctuations in producing SC.[21] Therefore, the enhancement of T_c in layered $CeMIn_5$ over $CeIn_3$ has been suggested to be due to their quasi-2D structure.[4,14,22] Our results suggest that the small window for the spin fluctuations regime in $CeIn_3$ may also be partly responsible for its lower T_c.

In conclusion, on the basis of the ^{115}In-NQRT_1 measurement, we have reported the P-induced evolution of the electronic and magnetic characteristics when approaching the SC phase in $CeRhIn_5$ and entering the SC phase in $CeIn_3$. In $CeRhIn_5$ that is already in the itinerant magnetic regime at $P=0$, T_N slightly increases with P at lower pressures which is in accordance with a previous report.[4] However, T_N starts to decrease above $P=1.23$ GPa approaching the critical value at which SC sets in. At the same time, a pseudogap behavior emerges. By contrast, in $CeIn_3$, the localized magnetic character is robust against the application of P up to 1.9 GPa, beyond which T^*, which marks an evolution into an itinerant magnetic regime, increases rapidly. It is interesting that the superconductivity emerges in such an itinerant regime. The window for the 3D AF fluctuation regime with respect to external pressure is much narrower in $CeIn_3$ than in $CeRhIn_5$, which may also be partly responsible for its lower T_c in $CeIn_3$.

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*Present address: Oxford Instruments, Tubney Woods, Abingdon, Oxon, OX13 5QX, UK.

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