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Enhancement of T_c in $CeIr(In_{1-x}Cd_x)_5$ studied by In-NQR

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Abstract. We report on superconducting nature under pressure in $CeIr(In_{1-x}Cd_x)_5$ by In-Nuclear-Quadrupole-Resonance (NQR) studies. In $CeIr(In_{0.925}Cd_{0.075})_5$, the inhomogeneous antiferromagnetic order at $T_N \sim 2.3$ K is induced by Cd-dopants and superconductivity disappears at ambient pressure. However, the measurements of a nuclear-spin-lattice-relaxation rate $1/T_1$ have revealed that the superconductivity suddenly occurs above 2.1 GPa. It is observed that the superconducting gap is enhanced in the Cd-doped sample, indicating that the Cd-doping induces the strong coupling superconductivity leading to the enhancement of T_c in the $CeIrIn_5$ system. Furthermore, we found that the residual density of states at the Fermi level increases with increasing pressure, suggesting that the superconducting nature of $CeIrIn_5$ is quite different from those of $CeCoIn_5$ and $CeRhIn_5$.

1. Introduction

The cerium (Ce)-based heavy-fermion compounds, $CeMIn_5$ (M = Co, Rh, and Ir) provide us with the opportunity to systematically investigate an interplay between antiferromagnetism (AF) and superconductivity (SC) [1, 2, 3, 4]. $CeIrIn_5$ and $CeCoIn_5$ show SC at ambient pressure (P) below $T_c = 0.4$ and 2.3 K, respectively [1, 2, 3, 4]. In $CeRhIn_5$, which is an antiferromagnet with $T_N = 3.8$ K at ambient P, SC occurs at $T_c \sim 2.2$ K under P over 2 GPa. Since the existence of two superconducting phases was reported in $CeRh_{1-y}Ir_yIn_5$ [5], it has been suggested that the superconducting nature in $CeIrIn_5$ is distinguished from those in $CeCoIn_5$ and $CeRhIn_5$. Figs.1(a) and 1(b) show the respective phase diagrams of SC (denoted as SC1 and SC2) for $CeRh_{1-y}Ir_yIn_5$ and $CeIrIn_5$ under P. SC1 is closely related to antiferromagnetic interactions, as argued extensively in $CeCoIn_5$[6] and $CeRhIn_5$[7], whereas it is reported from the previous NQR measurements that the SC2 in $CeIrIn_5$ occurs in a different situation from SC1 as follows [8]. The maximum of $T_c(= 0.9$ K) around 3 GPa takes place under the absence of antiferromagnetic correlations that is confirmed from the fact that the $(T_1T)^{-1}$-constant behavior is observed above T_c, where T_1 is a nuclear-spin-lattice-relaxation time. However, the values of $(T_1T)^{-1}$ still remain an order of magnitude larger than those in $LaIrIn_5$, indicating that some fluctuation except for AF spin fluctuations are dominant under high P. Actually, the large electronic specific heat

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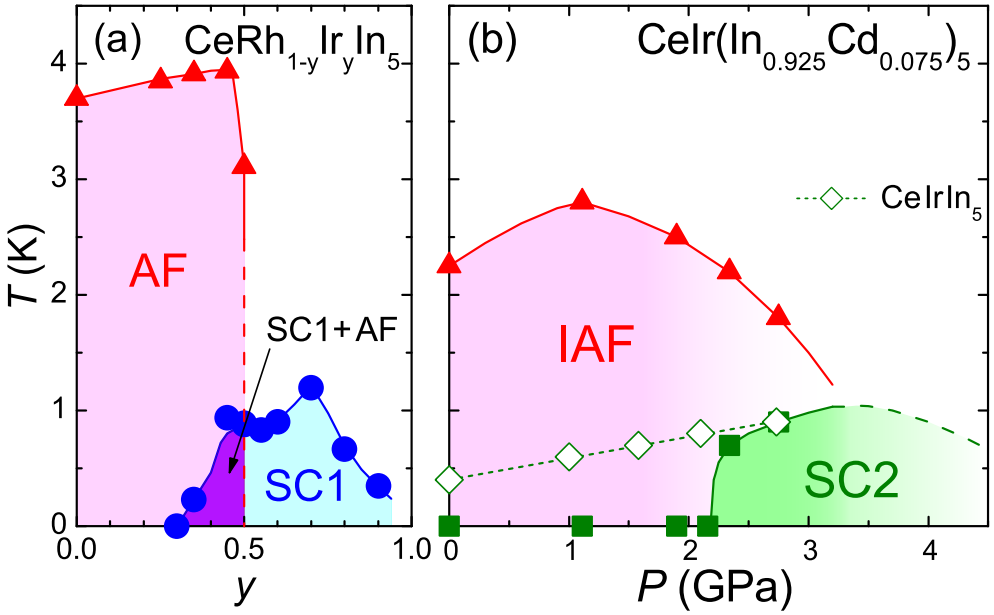


Figure 1. (Color online) (a) The phase diagram for $CeRh_{1-x}Ir_xIn_5$ as a function of Ir concentration. T_N and T_c are referred from Refs. [24, 25]. SC1 is expected to be mediated by magnetic correlations. (b) The $P-T$ phase diagram for $CeIr(In_{0.925}Cd_{0.075})_5$ partly from Ref. [26]. The T_N(solid) and T'_N(open triangles) are determined from the peak in the T dependence of $1/T_1$ and the broadening of NQR spectrum due to the onset of the antiferromagnetic order, respectively. The diamonds indicates T_c in pure $CeIrIn_5$ [8].

coefficient $\gamma \sim 0.38$ J/molK^2 at $P = 1.56$ GPa is observed in $CeIrIn_5$ [9]. Furthermore, from the resistivity measurements, a non-Fermi liquid behavior that the resistivity approximately follows a $T^{1.5}$ law has been observed in the wide P range up to 3.1 GPa [3].

Meanwhile, it was reported that all the anomalous transport properties observed in $CeRh_{0.2}Ir_{0.8}In_5$(SC1) and $CeIrIn_5$(SC2) originate from the AF spin fluctuations irrespective of the superconducting phase to which the system belongs. [10]. The detailed characteristics of AFM spin fluctuations including the dimensionality at ambient P in $CeIrIn_5$ are reported from the NMR measurements [11, 12]. In this context, the reason why the maximum T_c of SC2 in $CeIrIn_5$ is realized far away from an AF quantum critical point (QCP) is not clarified yet. The existence of two superconducting domes(SC1 and SC2) was first reported in $CeCu_2(Si_{1-x}Ge_x)_2$ [13]. It is suggested from extensive experimental and theoretical studies that the occurrence of SC2 in the high P region is mediated by valence fluctuations [14, 15, 16, 17]. In order to clarify the characters of SC2, we report the nuclear-quadrupaole-resonance (NQR) measurement under P in pure and Cd-doped $CeIrIn_5$.

Single crystals of $CeIr(In_{1-x}Cd_x)_5$ were grown by the In-flux method. They were crushed into coarse powder in order to allow RF pulses to easily penetrate the sample for NQR measurements. T_c's for the samples are determined by a sudden decrease in $1/T_1$ below T_c. Hydrostatic P was applied by utilizing a piston-cylinder cell(NiCrAl-BeCu double cylinder) filled with Daphne 7474 as a P-transmitting medium [18]. To calibrate P at low T's, the shift in T_c of Sn metal at P was monitored by resistivity measurements. $CeMIn_5$ consists of alternating layers of CeIn and MIn_4, and there are two sites—In(1) and In(2)—per unit cell. The In(1) and In(2) sites are located in the CeIn and MIn_4 layers, respectively. The measurements of T_1 were mainly performed at the transition of $2\nu_Q$ for the high symmetry In(1) site in $CeMIn_5$. T_1 was measured by the

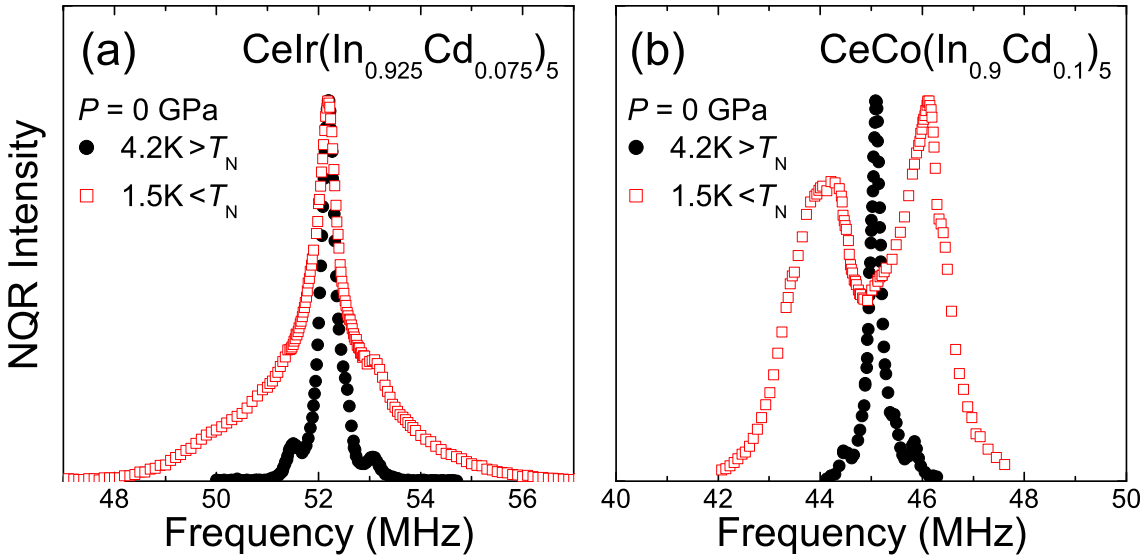


Figure 2. (Color online) (a) $3\nu_Q$ spectra at the In(2) site above and below $T_N \sim 2.3$ K in $CeIr(In_{0.925}Cd_{0.075})_5$. (b) $3\nu_Q$ spectra at the In(2) site above and below $T_N \sim 3$ K in $CeCo(In_{0.9}Cd_{0.1})_5$.

conventional saturation-recovery method. Here, an NQR frequency (ν_Q) is defined by the NQR Hamiltonian, $\mathcal{H}_Q = (h\nu_Q/6)[3{I_z}^2 - I(I+1) + \eta({I_x}^2 - {I_y}^2)]$, where η is the asymmetric parameter of the electric field gradient. $\eta = 0$ at the In(1) site is expected because of its good symmetry.

2. Experiments and discussion

Figure 1(b) shows the $P-T$ phase diagram in $CeIr(In_{0.925}Cd_{0.075})_5$. The antiferromagnetic order induced by the Cd-doping is observed in the wide P range. Fig. 2(a) shows the NQR spectra at the $3\nu_Q$ of the In(2) site above and below T_N at ambient P in $CeIr(In_{0.925}Cd_{0.075})_5$. The full width at half maximum (σ) of the paramagnetic NQR spectrum at $T = 4.2$ K is less than 350 kHz and considerably sharp, indicating the relatively high-quality of sample in spite of the Cd-doping as impurities. The main peak from ^{115}In nuclei is observed at ~ 52.2 MHz and the small peak at a little lower frequency is derived from the isotope ^{113}In. The origin of another peak at higher frequency than the main peak is not identified. Since it hardly broadens and is distinguishably observed below T_N in spite of no detection of the signal from ^{113}In, it may be associated with some impurity mixed into the sample. It is also possible that another peak at higher frequency than the main peak is derived from impurities induced by the Cd doping. The antiferromagnetic NQR spectrum at $T = 1.5$ K exhibits no clear splitting, but a large broadening at its tail in $CeIr(In_{0.925}Cd_{0.075})_5$. From the previous NQR measurements at the In(2) site in Cd-doped $CeCoIn_5$, as shown in Fig. 2(b), the clear splitting of the NQR spectrum is observed below T_N, indicating that the homogeneous antiferromagnetic order with a uniform magnetic moment M_{AF} over the whole sample is realized [19]. Therefore, the spectral shape below T_N in $CeIr(In_{0.925}Cd_{0.075})_5$ indicates the inhomogeneous antiferromagnetic order (IAF) with a large distribution of M_{AF}. The amplitude of M_{AF} may be locally enhanced near Cd-dopants through the reduction of hybridization between $4f$ and conduction electrons. The difference in the character of the AFM order is probably relevant to the fact that $CeCoIn_5$ is much closer to an AFM QCP than $CeIrIn_5$.

Next, we discuss superconducting characteristics for $CeIr(In_{1-x}Cd_x)_5$. As shown in Fig. 1(b), T_c is enhanced by the application of P in $CeIrIn_5$ in spite of moving the system further away from

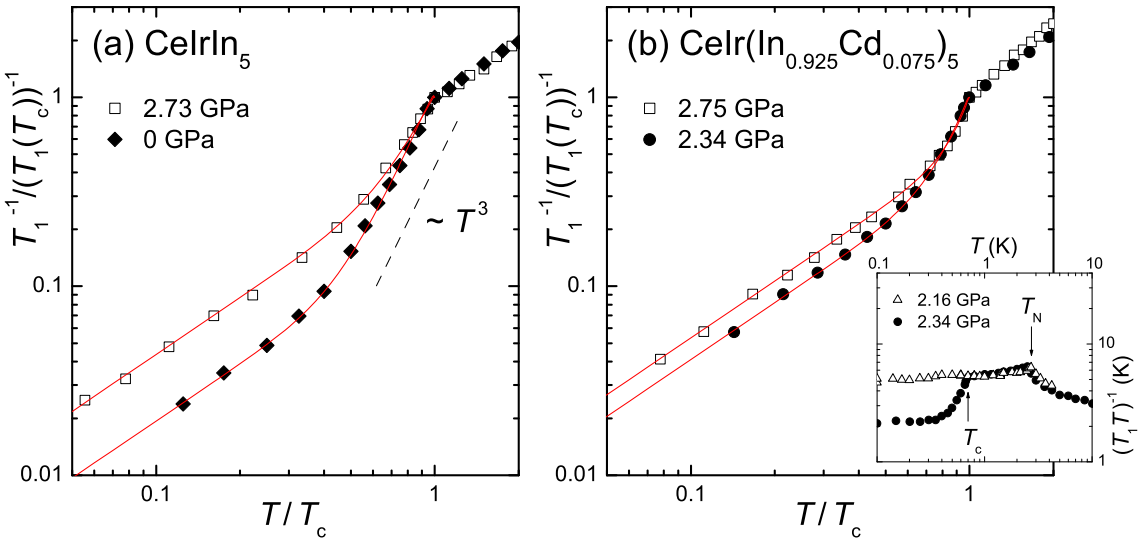


Figure 3. (Color online) (a) The T dependence of $1/T_1$ normalized at T_c at $P = 0$ and 2.73 GPa in $CeIrIn_5$. (b) The T dependences of $1/T_1$ normalized at T_c at $P = 2.34$ and 2.75 GPa in $CeIr(In_{0.925}Cd_{0.075})_5$. The inset is the T dependence of $(T_1T)^{-1}$ at $P = 2.16$ and 2.34 GPa. The solid lines in both figures are the calculated results (see text).

the antiferromagnetic QCP. The IAF phase is observed in wide P region in $CeIr(In_{0.925}Cd_{0.075})_5$. Therefore, SC is suppressed by the onset of the antiferromagnetic order and/or the impurity effect from Cd-dopants. The anomaly in the T dependence of $(T_1T)^{-1}$ at $T_N = 2.7$ K is derived from the onset of the IAF order at 2.16 GPa in the inset of Fig. 3, and no anomalies are found below T_N, indicating that SC does not occurs up to 2.16 GPa. When P is slightly more increased to 2.34 GPa, the rapid decrease of $(T_1T)^{-1}$ suddenly appears at 0.7 K, suggesting the occurrence of SC. The diamagnetism is also confirmed from the ac-susceptibility measurement using the in-situ NQR coil. Above 2.34 GPa, T_c continues to increase and reaches about 0.9 K, which is the maximum value of T_c around 2.8 GPa in pure $CeIrIn_5$.

The T dependences of $1/T_1$ normalized at T_c are shown for the pure and Cd-doped samples in Figs. 3(a) and (b), respectively. The T dependence of $1/T_1$ below T_c allows us to estimate a superconducting gap ($2\Delta_0/k_BT_c$) and a residual density of states (RDOS) at the Fermi level due to some impurity effect by assuming a certain pairing symmetry. Several models as a pairing symmetry in $CeIrIn_5$ have been suggested from experimental studies, but a $d_{x^2-y^2}$-wave model is recently supported by the thermal-conductivity, magnetic-penetration-depth, and specific-heat measurements [20, 21, 22]. Therefore, we tentatively assume a $d_{x^2-y^2}$-wave model to analyze $1/T_1$ data obtained from the present NQR measurements. The solid lines in Figs. 3 are the calculated results for $CeIrIn_5$ with parameters of ($2\Delta_0/k_BT_c$, RDOS) = (5.2, 0.44) at $P = 0$ and (5.2, 0.66) at $P = 2.73$ GPa. $2\Delta_0/k_BT_c$ is almost independent of P in $CeIrIn_5$, consistent with the previous NQR measurements [8]. The value of RDOS in $CeIrIn_5$ is fairly large. RDOS = 0.08 and 0.14 are confirmed at $P = 0$ in $CeCoIn_5$ [6] and at $P = 2.35$ GPa in $CeRhIn_5$ [23], respectively. Since the linewidth (σ) of the NQR spectrum at $2\nu_Q$ for the In(1) site in $CeIrIn_5$ is about 50 kHz, it is expected that the sample quality is considerably high. In the case of $CeCoIn_5$ and $CeRhIn_5$, $\sigma \sim 20$ and 50 kHz, respectively. Considering the existence of the large RDOS in spite of its high purity, it is possible that a superconducting gap is not formed or very small to be easily broken even by weak impurity scattering in one or a few bands of the Fermi surface. Here, we highlight the fact that RDOS in $CeIrIn_5$ increases from 0.44 to 0.66

despite the enhancement of T_c from 0.4 to 0.9 K. When T_c increases, it is simply expected that RDOS is reduced through the formation of a strong-coupling superconductivity. Nevertheless, RDOS is unexpectedly enhanced by the application of P. Such a behavior contrasts with those in $CeCoIn_5$ and $CeRhIn_5$ in which RDOS remains almost unchanged and is reduced as P increases, respectively. The unexpected enhancement of RDOS by the application of P cannot be understood in terms of a simple impurity effect. Furthermore, since $CeIrIn_5$ is probably farther from an AFM QCP than $CeCoIn_5$, the influence of antiferromagnetic spin fluctuations on RDOS should be small in $CeIrIn_5$, as compared with that in $CeCoIn_5$. In fact, antiferromagnetic spin fluctuations are greatly suppressed by the application of P in $CeIrIn_5$ [8]. The cause of the unexpected enhancement by P cannot be explained by the effect of antiferromagnetic spin fluctuation. Analogous SC characteristics are also observed in the Cd-doped sample with the parameters of ($2\Delta_0/k_BT_c$, RDOS) = (6.4, 0.64) and (7.7, 0.73) at P = 2.34 and 2.75 GPa, respectively, indicating that it is intrinsic in the $CeIr(In_{1-x}Cd_x)_5$ system. The value of RDOS in the Cd-doped sample is larger than that in the pure sample due to the impurity effect. It should be noted that $2\Delta_0/k_BT_c$ is enhanced by the Cd-doping in spite of the impurity effect and the induced IAF order. In order to solve the mechanism of SC2, it would be important to understand the role of the Cd-doping for SC2.

3. Conclusion

In conclusion, the present In-NQR measurements have revealed that the nature of SC2 in pure and Cd-doped $CeIrIn_5$ differs from that of SC1 that is closely related to the AFM correlation. In pure $CeIrIn_5$, the unexpected enhancement of RDOS by the application of P is observed in spite of no addition of impurity and the increase in T_c. The similar behavior is also observed in Cd-doped $CeIrIn_5$. The unexpected enhancement of RDOS may be related with the mechanism of SC2. The similar T_c to the maximum value($T_c^{max} \sim 0.9$ K) in pure $CeIrIn_5$ is realized even in Cd-doped $CeIrIn_5$ in spite of the existence of Cd impurities and the AF order. The Cd-doping induces the strong coupling superconductivity in the $CeIrIn_5$ system, leading to the enhancement of T_c for SC2. In order to understand the mechanism of SC2 in more detail, it is inevitable to clarify the cause for the unexpected enhancement of RDOS and T_c in the Cd-doped $CeIrIn_5$ system.

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