Enhancing the Superconducting Transition Temperature of the Heavy Fermion Compound $CeIrIn_5$ in the Absence of Spin Correlations

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We report on a pressure- (P-)induced evolution of superconductivity and spin correlations in $CeIrIn_5$ via the ^{115}In nuclear-spin-lattice-relaxation rate measurements. We find that applying pressure suppresses dramatically the antiferromagnetic fluctuations that are strong at ambient pressure. At $P = 2.1$ GPa, T_c increases to $T_c = 0.8$ K, which is twice T_c ($P = 0$ GPa), in the background of Fermi-liquid state. This is in sharp contrast to the previous case in which a negative, chemical pressure (replacing Ir with Rh) enhances magnetic interaction and increases T_c. Our results suggest that multiple mechanisms work to produce superconductivity in the same compound $CeIrIn_5$.

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The cerium- (Ce-)based heavy fermion compounds $CeMIn_5$ ($M =$ Co, Rh, and Ir) discovered a few years ago provide a unique opportunity to investigate the interplay between antiferromagnetism and superconductivity [1–3]. Among $CeMIn_5$, $CeIrIn_5$ and $CeCoIn_5$ show superconductivity at $P = 0$ below $T_c = 0.4$ and 2.3 K, respectively [2,3]. Antiferromagnet $CeRhIn_5$ becomes superconducting at relatively lower critical pressure $P_c \sim 1.6$ GPa and yet exhibits a higher $T_c \sim 2$ K [1]. Measurements of nuclear-quadrupole resonance (NQR) [4–7], thermal transport, and heat capacity [8] on $CeMIn_5$ found that the superconductivity is unconventional, with line nodes in the superconducting gap function. NQR [4,5,7,9] and inelastic neutron diffraction [10] measurements also found strong antiferromagnetic spin fluctuations in the normal state. In addition, in $CeIrIn_5$, the antiferromagnetic spin fluctuations are found to be anisotropic [4]; namely, a magnetic correlation length $\xi_{\rm plane}$ within the tetragonal plane grows more dominantly than ξ_c along the c axis associated with their two-dimensional crystal structure. The nuclear-spin-lattice-relaxation rate ($1/T_1$) was found to follow the relation of $1/T_1T \propto 1/(T + \theta)^{3/4}$ with a small value of $\theta =$ 8 K [4]. The same analysis was applied to $CeCoIn_5$ with resulting $\theta = 0.6$ K [9,11]. Note that θ is a measure as to what extent the system is close to an antiferromagnetic quantum critical point (QCP) [12]. It was suggested that the difference in the value of θ between $CeIrIn_5$ and $CeCoIn_5$ may lead to the large difference in the value of T_c.

Moreover, substituting Rh for Ir in $CeIrIn_5$ increases T_c up to 1 K in $CeRh_xIr_{1-x}In_5$ [13]. It was found that this substitution acts as negative chemical pressure that increases the antiferromagnetic correlations [14]. In fact, in $CeRh_{0.5}Ir_{0.5}In_5$, the enhanced superconductivity coexists microscopically with antiferromagnetic order that sets in at $T_N = 3$ K [14]. These results have naturally led to an expectation that superconductivity in $CeMIn_5$ is induced by antiferromagnetic correlations. However, it was found that applying hydrostatic pressure also increases T_c in $CeIrIn_5$ [15,16]. A $T_c^{\rm max} \sim 1$ K was found at around $P \sim 3$ GPa [15]. The possible role of magnetic correlations in the increase of T_c under pressure in $CeIrIn_5$ is still an open question.

In this Letter we report on the pressure-induced evolution of superconducting characteristics and antiferromagnetic spin fluctuations in $CeIrIn_5$ through T_1 measurements. We found that the superconductivity with enhanced T_c under pressure in $CeIrIn_5$ is realized in the absence of antiferromagnetic spin fluctuations. Our results suggest that there are two mechanisms for superconductivity in the same compound $CeIrIn_5$. We argue that the existence of multiple superconducting phase may be common in heavy fermion compounds.

Single crystals of $CeIrIn_5$ were grown by the self-flux method and moderately crushed into grains in order to allow rf pulses to penetrate easily into samples. To avoid crystal distortions, however, the grains' diameters were kept larger than 100 μm. $CeIrIn_5$ consists of alternating layers of $CeIn_3$ and $IrIn_2$ and hence has two inequivalent ^{115}In sites per unit cell. The ^{115}In-NQR measurements were made at the In(1) site [4] which is located on the top and bottom faces of the tetragonal unit cell. Since the position of the In(1) site is crystallographically closer to the Ce nucleus than that of the In(2) site, it is suited to investigate the relationship between superconductivity and magnetic correlations. ^{115}In-NQR measurement was made by a conventional saturation-recovery method. The ^{115}In-NQR 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. The value of pressure at low temperature was determined from the pressure dependence of the T_c value of Sn metal measured by a conventional four terminal method. For our pressure cells, the spatial distribution in values of pressure $\Delta P/P$ is estimated to be $\sim$3% from a broadening in the linewidth of NQR spectrum [17].

Figure 1 shows the T dependence of ^{115}In-NQR $1/T_1$ for $CeIrIn_5$ measured at $P = 0$, 1.0, 1.58, and 2.1 GPa. The data at $P = 0$ GPa and for $LaIrIn_5$ are taken from Ref. [4]. Above T_c the Ce $4f$ magnetic contribution to the relaxation rate for $CeIrIn_5$ is clear when comparing its value to $1/T_1$ measured in the nonmagnetic $LaIrIn_5$. As reported in a previous work, the sudden decrease in $1/T_1$ at $P = 0$ GPa at $T = 0.4$ K indicated the onset of bulk superconductivity. Unconventional superconductivity was evidenced from the characteristic T dependence of $1/T_1$ that exhibits no coherence peak just below T_c and follows the T^3 behavior well below T_c [4]. As pressure increases, T_c increases linearly and reaches $T_c = 0.8$ K, which is twice the T_c at $P = 0$ GPa. Note that the unconventional nature of superconductivity under pressure is evident from the T dependence of $1/T_1$ below T_c, as shown in Fig. 2.

In order to examine the pressure-induced evolution of superconducting characteristics in $CeIrIn_5$, $[T_1(T)^{-1}/T_1(T_c)^{-1}]$ versus $T/T_c(P)$ is plotted in Fig. 2. The line-node superconducting energy-gap model with $\Delta = \Delta_0 \cos\theta$ was applied to analyze the $1/T_1$ data below T_c

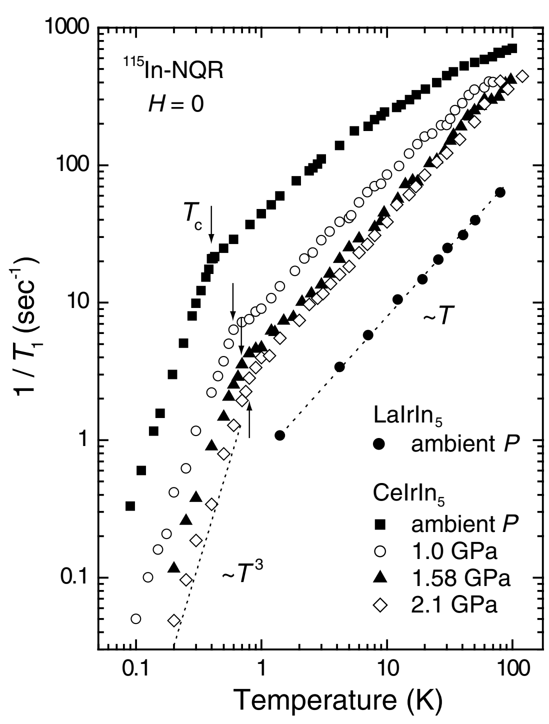

FIG. 1. The T dependence of $^{115}(1/T_1)$ in $CeIrIn_5$ at $P = 0$, 1.0, 1.58, and 2.1 GPa. The data for $CeIrIn_5$ at $P = 0$ GPa and $LaIrIn_5$ are taken from Ref. [4]. Arrows indicate a superconducting transition temperature T_c at each pressure. The respective dotted lines indicate the behaviors of $1/T_1T =$ const and $1/T_1 \propto T^3$ at the normal and superconducting state.

with Δ_0/k_BT_c as a parameter,

$$\frac{T_1(T_c)}{T_1} = \frac{2}{k_BT} \int \left(\frac{N_S(E)}{N_0}\right)^2 f(E)[1 - f(E)]dE,$$

where $N_S(E)/N_0 = E/\sqrt{E^2 - \Delta^2}$, with N_0 being the density of state in the normal state and $f(E)$ is the Fermi distribution function. From fittings shown by the solid line in Fig. 2, the pressure independent values of $\Delta_0/k_BT_c = 2.5$ are obtained. Here, we assumed the residual density of state in the superconducting gap to be zero since clear T^3 behavior is observed down to $0.15T_c$ in our sample. This result shows that the coupling strength for the formation of Cooper pairs is almost the same in $CeIrIn_5$ regardless of the increase in T_c. It is consistent with the results of specific heat measurements under pressure which showed that the relatively small specific heat jump at T_c, $\Delta C/\gamma(T_c) \sim 0.8$, is almost independent of pressure [16]. Here, γ is the T-linear coefficient in electronic specific heat.

What type of evolution in the electronic and magnetic properties under pressure increases T_c in $CeIrIn_5$? In order to gain insight into this issue, we focus on the pressure-induced evolution of magnetic fluctuations in the normal state in $CeIrIn_5$. Figure 3 and its inset show the $1/T_1T$ versus T plots in $CeIrIn_5$ at $P = 0$, 1.0, 1.58, and 2.1 GPa and $LaIrIn_5$ at $P = 0$ GPa in linear and logarithmic scales, respectively. At $P = 0$ GPa, the T dependence of $1/T_1T$ above T_c is well explained by the anisotropic antiferromagnetic spin-fluctuations model [4,12]. As seen in Fig. 3, the application of pressure markedly suppresses the antiferromagnetic spin fluctuations, bringing the system away from the antiferromagnetic QCP. As a result, a relation of

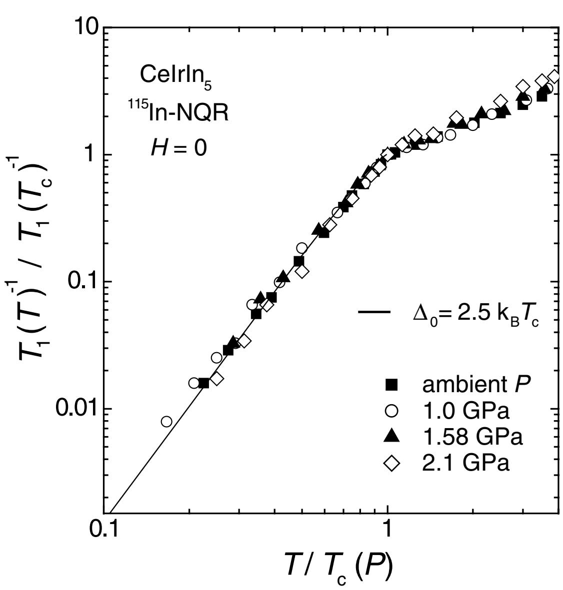

FIG. 2. Plot of $[T_1(T)^{-1}/T_1(T_c)^{-1}]$ versus $T/T_c(P)$. The solid line indicates a calculation based on an unconventional superconducting model with a line-node gap assuming $\Delta_0 = 2.5k_BT_c$ (see text).

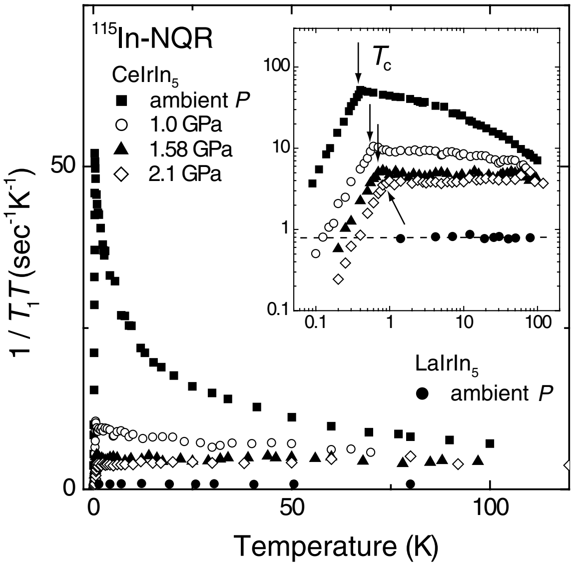

FIG. 3. The T dependence of $1/T_1T$ at $P = 0$, 1.0, 1.58, and 2.1 GPa. The data for $CeIrIn_5$ at $P = 0$ GPa and $LaIrIn_5$ are taken from Ref. [4]. In the main figure and the inset, the data are plotted in linear and logarithmic scales, respectively. Arrows and dotted lines indicate T_c (P) and a $T_1T = \text{const}$ relation for $LaIrIn_5$, respectively.

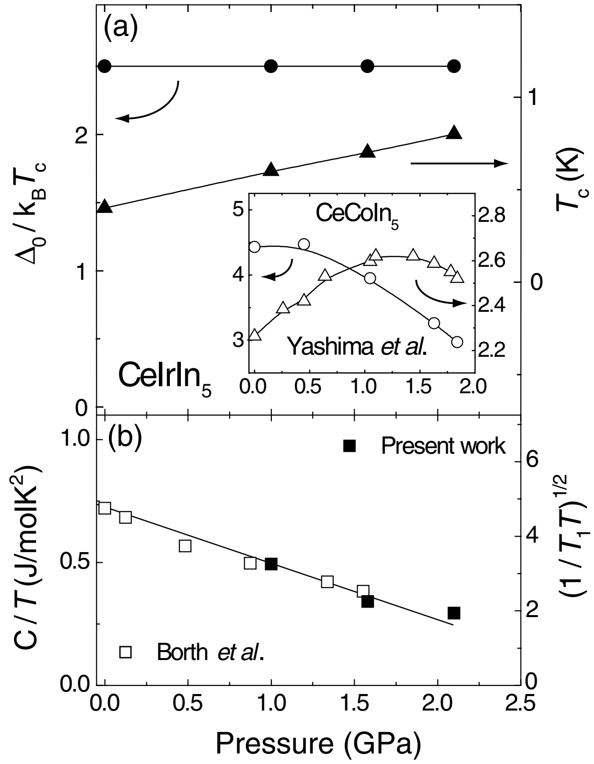

FIG. 4. (a) The pressure dependence of Δ_0/k_BT_c and T_c in $CeIrIn_5$. The inset indicates those for $CeCoIn_5$ taken from the Ref. [11]. (b) The pressure dependence of γ [16] and $(1/T_1T)^{1/2}$ in $CeIrIn_5$ just above T_c (P) (see text). Solid lines are guides to the eye. .

$1/T_1T = \text{const}$ becomes valid over $T = 1$–100 K at $P = 2.1$ GPa without the development of antiferromagnetic spin fluctuations upon cooling. On the other hand, it should be noted that, as seen in Fig. 4(b), the values of $1/T_1T$ and $\gamma \sim 0.25$ J/K^2 mol [16] for $CeIrIn_5$ at $P = 2.1$ GPa are 1 order of magnitude larger than that for $LaIrIn_5$. These results indicate that strong electron correlation still plays a central role in enhancing the values of $1/T_1T$ and γ, even though antiferromagnetic spin fluctuations disappear in $CeIrIn_5$ under pressure. This is in contrast to the case for $CeCoIn_5$ and $CeRhIn_5$ in which antiferromagnetic spin fluctuations enhance $1/T_1T$ upon cooling, even though each system is away from the antiferromagnetic QCP with applying pressure [6,11].

The pressure dependencies of Δ_0/k_BT_c and T_c for $CeIrIn_5$ are summarized in Fig. 4(a) together with those for $CeCoIn_5$ in the inset. In $CeCoIn_5$, the application of pressure also significantly suppresses $1/T_1$, which is dominated by antiferromagnetic spin fluctuations specific to the antiferromagnetic QCP. It shows very good agreement with specific heat measurements under pressure [18]. Although a jump in specific heat at T_c [$\Delta C/\gamma(T_c) \sim 5$] has a surprisingly large value at ambient pressure, indicative of a strong coupling superconductivity, this value shows a marked decrease against applying pressure [18]. Correspondingly, $1/T_1T$ is significantly suppressed as pressure increases [11]. Both results suggest that the application of pressure to $CeCoIn_5$ increases the heavy-fermion bandwidth due to the increase of hybridization between f electrons and conduction electrons and eventually brings the system away from the antiferromagnetic QCP. As a result, the superconducting gap or Δ_0/k_BT_c in $CeCoIn_5$ decreases [11,18], as shown in the inset of Fig. 4(a). Noting that, in $CeCoIn_5$, the $1/T_1T$ is intimately enhanced upon cooling to T_c, it is expected that antiferromagnetic spin fluctuations play a role in mediating the Cooper pairs even when the system is away from the antiferromagnetic QCP under pressure [11]. Nevertheless, the enhancement of T_c with applying pressure was suggested to be relevant to the increase in heavy-fermion bandwidth that is expected to make the lifetime of quasiparticles long enough. In this context, the Cooper pairs in $CeCoIn_5$ may originate from attractive interaction induced by antiferromagnetic spin fluctuations. It was hence argued that, in the presence of antiferromagnetic spin fluctuations, the value of T_c may be controlled by the combined effect of coupling strength for the Cooper-pair formation due to the closeness to the antiferromagnetic QCP and the heavy-fermion bandwidth in $CeCoIn_5$ [11].

This approach fails to account for the pressure dependence of T_c in $CeIrIn_5$, however. Indeed, we have shown that the maximum of T_c is realized without the development of antiferromagnetic spin fluctuations in the normal state as supported by the observation of $T_1T = \text{const}$ law over two decades in the T range above T_c. The heavy-fermion bandwidth increases with pressure as corroborated by the fact that γ [16] is scaled to $(1/T_1T)^{1/2}$, as seen in Fig. 4(b). Here, the value of $(1/T_1T)^{1/2}$ is proportional to the density of states at the Fermi level. Therefore, the increase of T_c may be relevant to the increase of heavy-fermion bandwidth.

In most Ce-based heavy fermion compounds, where the superconductivity appears either at $P = 0$ GPa or in the neighborhood of antiferromagnetism, the antiferromagnetic spin fluctuations are expected to be responsible for the onset of unconventional spin-singlet superconductivity. In the antiferromagnetic spin-fluctuations theory [19], T_c takes a maximum value at the border of antiferromagnetism as observed in previous examples. In $CeCu_2X_2$ ($X =$ Si and Ge), however, the maximum value of T_c appears far away from the antiferromagnetic QCP, which indicates that the low-lying antiferromagnetic spin fluctuations are not responsible for the formation of superconductivity [20]. In fact, it has been found that two superconducting phases exist in $CeCu_2(Si_{1-x}Ge_x)_2$ [21]. It has been suggested that one of superconductivity (SC-I) is induced by strong antiferromagnetic spin fluctuations on the verge of antiferromagnetism and the other (SC-II) by valence instability of localized Ce $4f$ electrons since the system is far away from the antiferromagnetic QCP [20,22,23]. Markedly, the higher T_c takes place in SC-II.

Approaches based on spin-fluctuation theories also fail to account for the existence of spin-triplet superconductivity in Sr_2RuO_4 where two-dimensional Fermi-liquid state is realized with the strong electron correlation as confirmed by the $T_1T =$ const law. It has been proposed that the on-site electron correlation induces various types of unconventional superconductivity through the momentum dependence of quasiparticle interaction, which originates from the many-body effect [24]. Interestingly, this scenario can predict d-wave and p-wave superconductivity near half-filling and away from half-filling, respectively, without involving spin fluctuations as the key mechanism for pairing [24].

An important fact revealed by the present experiment is that superconductivity in $CeIrIn_5$ is robust over a wide pressure range where the antiferromagnetic spin fluctuations are absent. This may be due to either charge valence instability or on-site Coulomb interactions. Further experiments are required at this stage to clarify this issue.

In summary, we have presented the unique characteristics of superconductivity and its relation to antiferromagnetic spin fluctuations in the heavy-fermion superconductor $CeIrIn_5$ through ^{115}In-NQR measurements under pressure. The application of external pressure rapidly suppresses antiferromagnetic spin fluctuations that are strong at ambient pressure. At $P = 2.1$ GPa, where a $T_1T =$ const law is valid over $T = 1$–100 K, T_c increases up to $T_c = 0.8$ K, which is twice the T_c at $P = 0$ GPa. Our results indicate that another superconducting phase exists in the absence of antiferromagnetic spin fluctuations, in addition to the superconducting phase with $T_c^{\max} = 1$ K that coexists with antiferromagnetism. The present system bears some similarity with another prototype heavy fermion compound $CeCu_2Si_2$, and suggests that the existence of multiple superconducting phase may be common in heavy fermion compounds.

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