NMR and NQR studies of the heavy fermion superconductors CeTIn$_5$ (T=Co and Ir)

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We have carried out ^{115}In and ^{59}Co nuclear quadrupole resonance and nuclear magnetic resonance measurements on CeCoIn$_5$ and CeIrIn$_5$. The temperature T dependence of the nuclear spin-lattice relaxation rate $1/T_1$ of ^{115}In in the normal state indicates that CeCoIn$_5$ is located just at an antiferromagnetic instability, and CeIrIn$_5$ is in the nearly antiferromagnetic region. In the superconducting state, $1/T_1$ has no Hebel-Slichter coherence peak just below T_C and a power-law T dependence (close to T^3) at very low temperatures, which indicates the existence of line nodes in the superconducting energy gap. The ^{115}In (Ce-In plane) Knight shift in CeCoIn$_5$ decreases for both parallel and perpendicular directions to the tetragonal c axis in the superconducting state, which shows that the spin susceptibility decreases in all directions. These results indicate that CeCoIn$_5$ and CeIrIn$_5$ exhibit non-s-wave even parity (probably d-wave) superconductivity.

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I. INTRODUCTION

The occurrence of superconductivity in strongly correlated f-electron systems has intrigued researchers for more than two decades. In these systems, the hybridization between the conduction electrons and the localized f electrons results in the formation of heavy quasiparticles and leads to ferromagnetic (F) or antiferromagnetic (AF) spin fluctuations.[1] Since heavy fermion superconductors are located near the F or AF region, the existence of spin fluctuations has led to the prediction that superconductivity with non-s-wave symmetry, mediated by magnetic electron-electron coupling, is realized in heavy fermion systems.[2] More than five years ago, CeCu$_2$Si$_2$ (Ref. 3) and CeCu$_2$Ge$_2$ (at high pressure)[4] were the only superconducting members of the 4f-heavy fermion class. Since 1995, however, CePd$_2$Si$_2$,[5,6] CeRh$_2$Si$_2$,[7] CeIn$_3$,[8] and CeNi$_2$Ge$_2$ (Ref. 9) have been shown to become superconducting under pressure (P), which all have very low superconducting transition temperatures (T_C) at high pressure. Owing to the severe experimental conditions, little knowledge of the superconductivity was obtained in these systems. Recently, another pressure induced superconductor CeRhIn$_5$, which has the tetragonal HoCoGa$_5$ structure, has been discovered.[10] The ground state is AF for $P<16$ kbar, and superconductivity occurs at $T_C=2.2$ K for $P>16$ kbar. The ^{115}In nuclear quadrupole resonance[11] and the neutron diffraction[12] measurements determined the spiral spin structure of CeRhIn$_5$. Following the discovery of CeRhIn$_5$, two new isostructural superconductors CeIrIn$_5$ (Ref. 13) and CeCoIn$_5$,[14] were observed at ambient pressure. The respective values of T_C of the two compounds are 0.4 and 2.3 K. Superconductivity occurs at relatively high temperatures for the CeTIn$_5$ (T=Co, Rh, and Ir) compounds,[15] which makes them very suitable for microscopic measurements.

In CeIrIn$_5$ and CeCoIn$_5$, the electrical resistivity passes through a maximum around 50 K that typically is attributed to the cross-over from incoherent scattering of conduction electron at high T to the development of correlated bands at low T. Below about 20 K, the resistivity of both systems does not have a quadratic T dependence. The resistivity varies as $\rho=\rho_0+aT^n$ with n = 1.3 in CeIrIn$_5$ and n = 1 in CeCoIn$_5$, which are not described by the Landau Fermi liquid picture. The electronic specific heat coefficients γ at very low T in these systems are large, 750 mJ/mole K^2 for CeIrIn$_5$ and 350 mJ/mole K^2 for CeCoIn$_5$. In the stoichiometric compound, deviation from the Landau Fermi liquid picture, the "non-Fermi-liquid state," occurs when the system is located in the vicinity of a quantum critical point (around magnetic instabilities) and thermal and transport properties are affected by strong spin fluctuations.

Nuclear magnetic resonance (NMR) and nuclear quadrupole resonance (NQR) are powerful tools for studying magnetism and superconductivity, i.e., the spectrum and nuclear spin lattice relaxation rate $1/T_1$ provide valuable information about the magnetic structure, the spin fluctuations and, also, the symmetry of the superconductivity. The results of such a study of CeRhIn$_5$ have already been published and reveal anisotropic superconductivity with line nodes that occurs near the AF state.[16] In order to understand the interplay of spin fluctuations and superconductivity, NQR and NMR measurements on CeIrIn$_5$ and CeCoIn$_5$ have been performed. In CeTIn$_5$, there are two crystallographically nonequivalent In sites. One site is located in the Ce-In plane. This In site is surrounded by Ce ions and has a symmetry which is invariant under fourfold rotations about the c axis (hereafter referred to as site A in this paper). The other site is

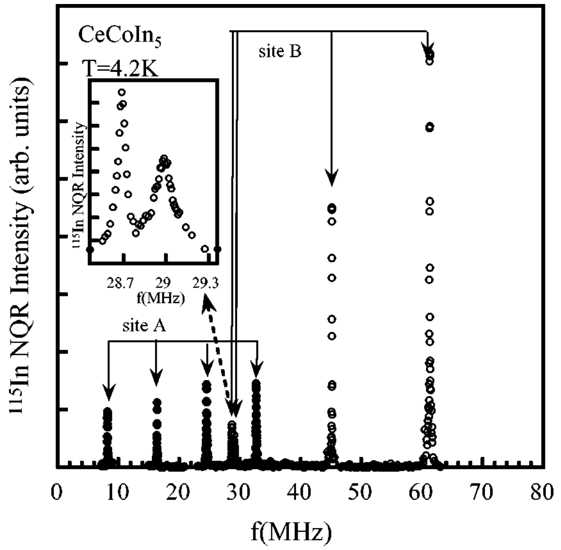

FIG. 1. ^{115}In NQR spectra obtained in CeCoIn$_5$. The closed circles represent the signals from site *A*, and open circles represent those from site *B*. The two lines from site *B* are nearly coincident at 29 MHz, which can be seen clearly in the inset.

surrounded by Ce and Co (or Ir) ions and has a lower crystal symmetry (referred to as site B). Due to the noncubic environment, one expects a large electric field gradient for both In sites, which is favorable for observing distinct NQR lines. In this paper, we report ^{115}In and ^{59}Co NMR/NQR studies in CeIrIn$_5$ and CeCoIn$_5$ at temperatures down to 50 mK.

II. EXPERIMENTAL

The ^{115}In NQR measurements were performed using a phase-coherent pulsed NMR/NQR spectrometer in the resonance frequency range 5-90MHz. Field-swept ^{115}In and ^{59}Co NMR spectra were obtained using a superconducting magnet with a maximum magnetic field of 50 kOe. Single crystals of CeCoIn$_5$ and CeIrIn$_5$ were grown from an In flux and crushed into powders for NQR and NMR measurements. The NMR measurements were carried out on aligned powders, in which the grains were aligned with the *c* axis parallel to magnetic field $H_{\rm ext}$. Two pieces of CeCoIn$_5$ crystals which were not crushed, were used to study the directional variation of K with respect to $H_{\rm ext}$. The ^{59}Co Knight shift ^{59}K was determined with respect to the ^{59}Co resonance in K$_3$Co(CN)$_6$, and the ^{115}In Knight shift ^{115}K with respect to the ^{115}In resonance in InCl$_3$. The value of T_1 was obtained by the recovery of the nuclear magnetization $m(t)$ after a saturation pulse. The measurements above 1.3 K were performed using a ^{4}He cryostat, and below 1.3 K with a ^{3}He cryostat and a ^{3}He-^{4}He dilution refrigerator.

III. RESULTS

A. ^{115}In NQR spectra

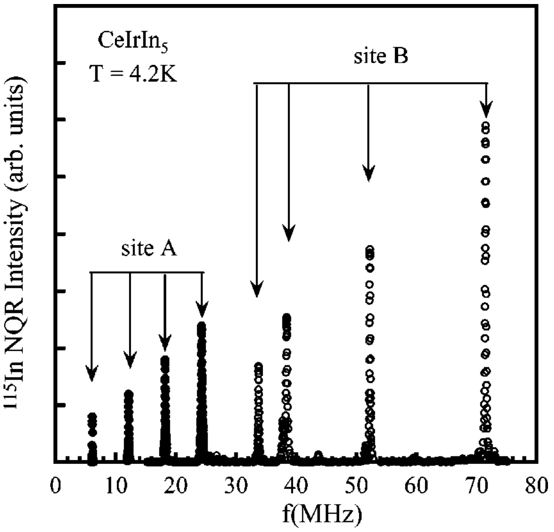

FIG. 2. ^{115}In NQR spectra obtained in CeIrIn$_5$.

Figures 1 and 2 show the ^{115}In NQR spectra obtained in CeCoIn$_5$ and CeIrIn$_5$ at $T=4.2$ K. The respective spectra consist of 8 narrow resonance lines, which could be assigned as the signals arising from two In sites. The electric quadrupole Hamiltonian is written as

$$\mathcal{H}_Q=\frac{e^2qQ}{4I(2I-1)}\left[3I_z^2-I(I+1)+\frac{\eta}{2}(I^{+2}+I^{-2})\right].$$

Here Q represents the nuclear quadrupole moment, the electric-field gradients are contained in eq and the asymmetric parameter η, defined as $eq\equiv V_{ZZ}$ and $\eta\equiv(V_{XX}-V_{YY})/V_{ZZ}$. By convention, V_{ZZ} has the largest magnitude, and V_{XX} and V_{YY} are chosen so that $0\leqslant\eta\leqslant1$. For $I=9/2$ (^{115}In), the four "allowed" transitions would be observed when η is small. The resonace frequencies were estimated by diagonalizing the electric quadrupole Hamiltonian. In the case of $\eta=0$, a set of lines would be observed at the resonance frequencies of $\nu_Q\equiv3e^2qQ/2I(2I-1)h$, $2\nu_Q$, $3\nu_Q$ and $4\nu_Q$, where h represents Planck constant. Indeed, a set of lines observed at 8.17, 16.34, 24.51, and 32.68 MHz in CeCoIn$_5$ are equally separated, which represents $\eta=0$. Similar signals are also observed in CeIrIn$_5$ at resonance frequencies of 6.07, 12.13, 18.20, and 24.27 MHz. These signals arise from the site A (In atoms in the Ce-In plane) which has an axially symmetric electric field gradient. From the crystal structure, it is evident that the direction of the electric field gradient V_{ZZ} is parallel to the tetragonal *c* axis. The values of the electric field gradient were evaluated as $e^2qQ/h=196$ MHz for CeCoIn$_5$ and 146 MHz for CeIrIn$_5$, respectively. Not only the four signals from site *A*, but also other signals are observed at 28.65, 28.98, 45.08, 61.21 MHz in CeCoIn$_5$, and 33.70, 38.35, 52.19, 71.43 MHz in CeIrIn$_5$. These signals arise from the site *B*, in which the ^{115}In nuclei are surrounded by Ce and Co (or Ir) ions. The corresponding electric field gradients are evaluated as $e^2qQ/h=372$ MHz with $\eta=0.39$ for CeCoIn$_5$, and $e^2qQ/h=436$ MHz with $\eta=0.46$ for CeIrIn$_5$. In either case, the associated "forbidden" transition ($|\Delta m|>1$), which exists in the case of finite η, was not visible. Since the symmetry of the electric field gradient in this site is lower than uniaxial, the principal axes of the tensor could not be determined only from NQR. Any

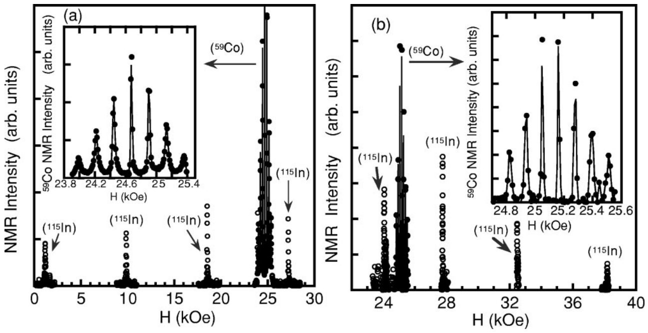

FIG. 3. ^{115}In and ^{59}Co NMR spectra measured at 4.2K in CeCoIn$_5$. (a) and (b) were obtained with the *c*-axis parallel and perpendicular to H_{ext}, respectively.

change of the spectra were not observed down to 50 mK in both compounds. It is noteworthy that the whole spectra could be explained by only the electric quadrupole interaction, which shows both compounds are in the paramagnetic state.

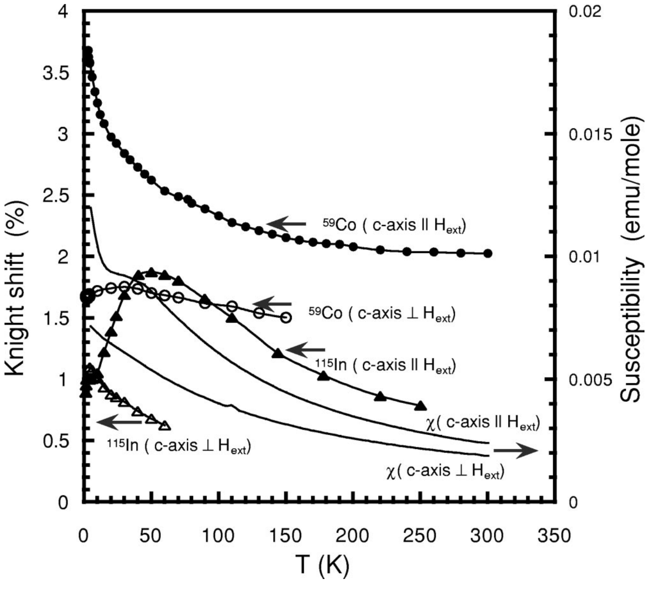

FIG. 4. Temperature dependence of the magnetic susceptibility and Knight shifts ^{59}K and ^{115}K in CeCoIn$_5$. The anisotropy of *K* is larger than that of χ.

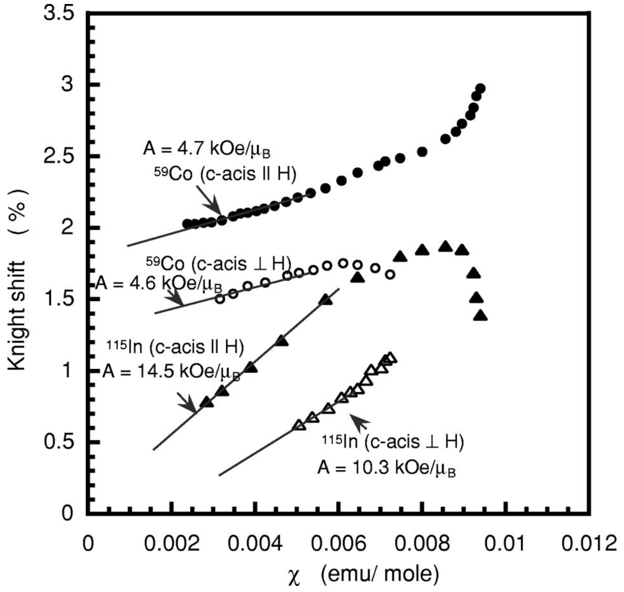

FIG. 5. Clogston-Jaccarino plot for ^{59}K and ^{115}K versus χ in CeCoIn$_5$.

B. ^{115}In and ^{59}Co NMR spectra in CeCoIn$_5$

The ^{115}In and ^{59}Co NMR were measured in CeCoIn$_5$ at 25.600 MHz and 4.2 K. Displayed in Figs. 3(a) and 3(b) are spectra obtained with the *c* axis parallel and perpendicular to H_{ext}, respectively. A set of widely separated ^{115}In NMR lines were observed which was assigned as the signal from site A, indicating a huge electric field gradient at the In site. In addition to ^{115}In NMR from site *A*, ^{59}Co NMR was also observed, which showed the direction of $V_{ZZ}(^{59}\text{Co})$ to be the *c* axis with the magnitude of $e^2qQ/h=1.58$ MHz and $\eta=0$. From these spectra, the parallel ($\|$) and perpendicular ($\perp$) components of ^{115}K and ^{59}K with respect to the *c* axis, whose *T* dependences are shown in Fig. 4, were evaluated. These Knight shifts *K* were compared with the magnetic susceptibility χ of CeCoIn$_5$ which is also plotted in Fig. 4. The presence of crystalline electric field that lifts the degeneracy of $J=5/2$ multiplet and induces the large anisotopy in the magnetic susceptibility and *K*. It is remarkable that ^{115}K$_\|$ has a broad maximum around 50 K, even if the corresponding anomaly in $\chi_\|$ is much smaller. The band structure calculations for CeIrIn$_5$ indicates that the Fermi surface is produced mainly by $5d$ and $4f$ electrons of Ce and $5p$ electrons of In (site *A*).[17,18] Hence, there is a strong coupling of electrons in the Ce-In plane. On the contrary, the hybridization of $5d$ electrons of Ir and $5p$ electrons of In at the *B* site makes bonding and antibonding bands, which results in a small density of states around the Fermi level E_F at Ir (Co). In some compounds containing unstable-moments, such as CeSn$_3$ and YbCuAl, the linear relation of χ and the Knight shift breaks down below T_{max}, which reflects the modification of the electronic structure associated with the formation of $4f$-ion coherence.[19,20] Although the modification of the electronic state is usually small in heavy fermion compounds, one explanation for the anomaly in *K* is the occurrence of reformation of the band structure below T_{max}. For the perpendicular component, no anomaly was observed in $^{115}K_\perp$, which increases monotonicallly with decreasing *T*. Figure 5 gives a Clogston-Jaccarino plot of Knight shift *K* versus susceptibility per mole Ce, with temperature an implicit parameter. A linear relation between K and χ is observed in the parallel component above 90 K with the coupling constant of 14.5 kOe/μ_B, which is nearly comparable with the coupling constant of the perpendicular component of 10.3 kOe/μ_B. Presumably reflecting the small coupling of the Ce-In plane and Co at the interlayer site, the *T* dependence of ^{59}K$_\|$, which is different from ^{115}K$_\|$, increases monotonically with decreasing *T*. ^{59}K$_\|$ is *T* dependent and is positive, which is induced by antiparallel Co $3d$ spins through $3d$-core polarization. The positive and nearly *T* independent ^{59}K$_\perp$ reveals

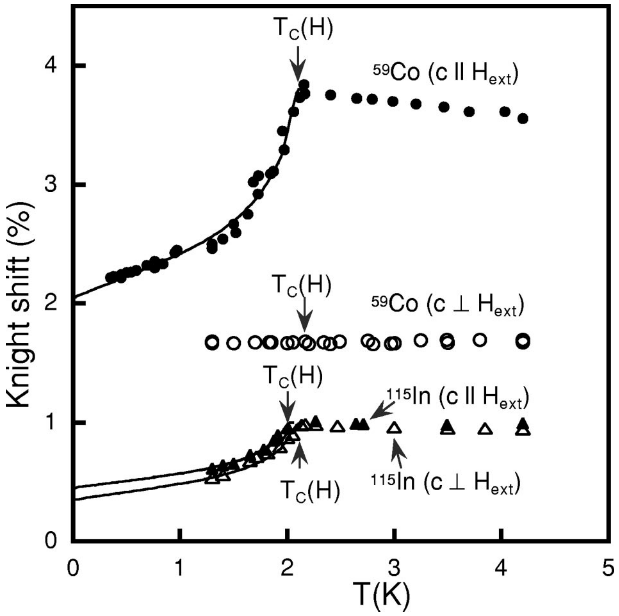

FIG. 6. T dependence of K in the superconducting state of CeCoIn$_5$.

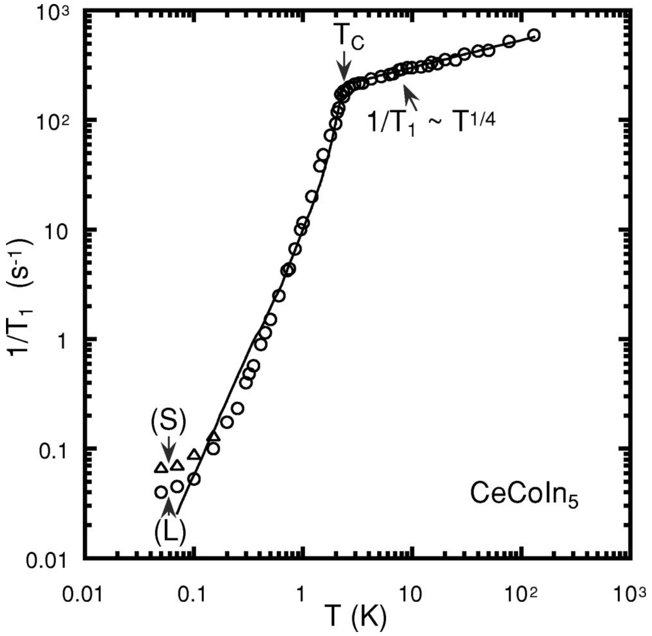

FIG. 7. T dependence of $1/T_1$ in CeCoIn$_5$. The values of $1/T_1$ below 1.3 K and above 1.3 K were evaluated from the $\pm 1/2 \leftrightarrow \pm 3/2$ and $\pm 7/2 \leftrightarrow \pm 9/2$ transitions, respectively. $1/T_1$ evaluated with the polar model is shown as the solid line. Below 0.2 K, the recovery curves could not be fitted by a unique value of $1/T_1$. The shorter components are plotted as open triangles.

that the shift is almost determined by the Co orbital contribution. A Clogston-Jaccarino plot for ^{59}K is also shown in Fig. 5. The evaluated coupling constants are 4.7 kOe/μ_B for $^{59}K_{\parallel}$ and 4.6 kOe/μ_B for $^{59}K_{\perp}$.

As seen in Fig. 6, a large decrease of ^{115}K was observed in both directions below T_C. For ^{59}K, the decrease was observed only in the parallel component, and not in the perpendicular component. It is expected that no decrease of $^{59}K_{\perp}$ reflects large T-independent Co $3d$ orbital contribution and small transferred contribution from Ce. The Ce susceptibility has also van Vleck component $\chi_{vv,\alpha\beta} \propto \Sigma_n[(0|J_\alpha|n)\times(n|J_\beta|0)/(E_n-E_0)]$, where $|0)$ and $|n)$ is the ground and excited states splitted by the crystalline electric field. It is noted that $\chi_{vv,\alpha\beta}$ contains spin part and induces spin polarization at the Co site, which does not decrease in the superconducting state.

C. nuclear spin-lattice relaxation rate $1/T_1$

Using ^{115}In NQR at site A, the nuclear spin lattice relaxation rates in both systems were measured. The expected functional form for $m(t)$ for spin $I=9/2$ and $\eta=0$ is a sum of four exponents with the coefficients determined by the initial conditions.[21] The function $m(t)$ contained only two fitting parameters $1/T_1$ and the numerical factor C, which is given by

$$1-\frac{m(t)}{m(\infty)}=C\left[b_1\exp\left(-\frac{3t}{T_1}\right)+b_2\exp\left(-\frac{10t}{T_1}\right)+b_3\exp\left(-\frac{21t}{T_1}\right)+b_4\exp\left(-\frac{36t}{T_1}\right)\right].$$

In our measurements, $1/T_1$ was obtained by different quadrupole transitions $\pm 1/2 \leftrightarrow \pm 3/2$, $\pm 3/2 \leftrightarrow \pm 5/2$, and $\pm 7/2 \leftrightarrow \pm 9/2$. The corresponding b- coefficients are described in Ref. 21. The T dependence of the $1/T_1$'s obtained in both compounds are shown in Figs. 7 and 8. In CeCoIn$_5$, the T dependence is close to $T^{1/4}$. In CeIrIn$_5$, $1/T_1$ in the normal state varies nearly as $T^{1/2}$ in the temperature range

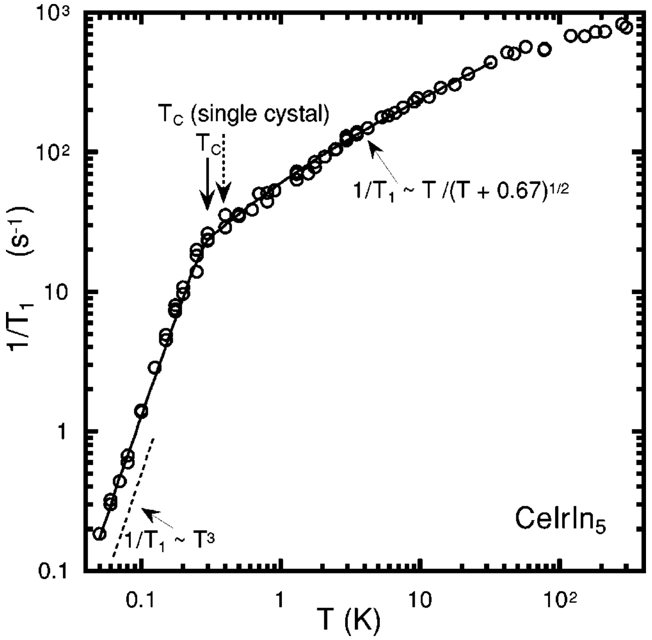

FIG. 8. T dependence of $1/T_1$ in CeIrIn$_5$. The values of $1/T_1$ below 1.3, between 1.3 and 4.2 K, and above 4.2 K were evaluated from the $\pm 1/2 \leftrightarrow \pm 3/2$, $\pm 3/2 \leftrightarrow \pm 5/2$, and $\pm 7/2 \leftrightarrow \pm 9/2$ transitions, respectively.

between 10 and 80 K, and the T dependence increases gradually below about 10 K with decreasing T. $1/T_1$ is expected to be proportional to the temperature in a model of simple Fermi-liquid behavior, which was not observed in both compounds. $1/T_1$ decreases rapidly just below $T_C = 2.3$ K in CeCoIn$_5$, and below about 0.3 K, which is lower than $T_C = 0.4$ K of a single crystal, in CeIrIn$_5$. Since NQR measurements were performed on a powder, T_C might decrease due to defects induced by crushing of the single crystal. It is expected that compounds having a lower T_C and a larger superconducting coherence length would be more sensitive to defects. In both compounds, Hebel-Slichter coherence peaks were not observed. In CeIrIn$_5$, $1/T_1$ varies nearly proportionally to T^3 at low temperatures. In CeCoIn$_5$, however, the T dependence becomes saturated below 0.3 K, where the recovery curve was not fitted by a unique T_1. The shorter and longer components of $1/T_1$ are shown in Fig. 7. This saturation would be caused by the extrinsic contribution to T_1 from a small amount of paramagnetic impurities, which masks the intrinsic superconducting quasiparticle contribution.

IV. DISCUSSION

It was found recently that AF ordering appears in Ce(Ir$_x$Rh$_{1-x}$)In$_5$ for $x < 0.6$ (Ref. 22) and in Ce(CoxRh$_{1-x}$)In$_5$ for $x < 0.7$.[23] Also the T dependence of the resistivity in CeCoIn$_5$ and CeIrIn$_5$ are not described by the Landau Fermi-liquid picture.[13,14] These results suggest that CeIrIn$_5$ and CeCoIn$_5$ are located close to AF state. Indeed, the observed T dependence of $1/T_1$ in CeCoIn$_5$ and CeIrIn$_5$ differs from the issue expected in the simple Fermi-liquid state, i.e., $1/T_1$ varies in proportion to T. $1/T_1$ would be governed by the strong AF spin fluctuations. In this case, one way to analyze $1/T_1$ is the use of the spin fluctuation theory developed by Moriya, which shows $1/(T_1T) \propto \sqrt{\chi_Q}$ around antiferromagnetic instabilities in the three-dimensional case. Here χ_Q represents the staggered susceptibility. In the theory,[24] χ_Q is determined by four parameters, which, however, were not yet determined in CeTIn$_5$. Therefore, we simply assume Curie-Weiss behavior for χ_Q, which is similar to the $3d$ transition metal case,[25]

$$\chi_Q \propto \frac{1}{T+\theta}.$$

In this model, the observed T dependence of $1/T_1$ (close to $\sqrt{T}$) in CeIrIn$_5$ was explained by taking the value of $\theta \sim 0.67$. The spin fluctuation theory predicts a $T^{1/4}$ dependence of $1/T_1$ near an AF instability, which was observed in CeCoIn$_5$.

Next, the ratio of $1/T_1$ in the superconducting state and that of the normal state, T_{1N}/T_{1S}, is related to the density of states (DOS) $N_S(E)$ as

$$\frac{T_{1N}}{T_{1S}} = \frac{2}{k_BT}\int_0^\infty [N_S(E)^2 + M(E)^2]f(E)[1-f(E)]dE,$$

where $M(E)$ is the "anomalous DOS" arising from the coherence effect which only exists in s-wave superconductivity, and $f(E)$ is the Fermi distribution function. $1/T_{1N}$ is evaluated by extending the normal state data from high temperatures. Unconventional superconductivity is characterized by the superconducting gap structure which has nodes along certain direction. Generally, energy gap has points or line nodes, which satisfy the group theoretical restriction arising from the crystal symmetry. In most of the case, the detailed structure of the gap function is remaining as an unresolved issue. However, a crude evaluation of $1/T_1$ would be possible by the simple polar model $\Delta = \Delta_0 \cos\theta$, since $1/T_1$ would likely be insensitive to points nodes in the presence of line nodes. If we calculate T_1, tentatively assuming $2\Delta_0(0) = 10\ k_BT_C$ for CeCoIn$_5$ and $5\ k_BT_C$ for CeIrIn$_5$, the experimental values are reproduced well as plotted in Figs. 7 and 8. The nearly T^3 law of $1/T_1$ is reminiscent of the relaxation behavior in many heavy fermion superconductors CeCu$_2$Si$_2$,[26] UPt$_3$,[27] UBe$_{13}$,[28] URu$_2$Si$_2$,[29] and UPd$_2$Al$_3$.[30]

By using the same energy gap, the T dependence of K is also analyzed in the case of even parity

$$K_S \propto \chi_S \propto \int N_S(E)\frac{df(E)}{dE}dE,$$

which is also plotted as the solid lines in Fig. 6. The calculation has reproduced the T dependence of the observed Knight shift. One of the aspects of K in the superconducting state is that K varies nearly linearly with T at very low temperatures, which arises from the low-energy excitation of quasiparticles in the superconducting state. The existence of line nodes explains the T dependence. The fact that K and $1/T_1$ were consistently explained by an anisotropic spin singlet model, strongly indicates the appearance of anisotropic even parity (probably d wave) superconductivity in CeTIn$_5$. Further measurements using a directional probe in k space, such as thermal conductivity, would be useful for determining the precise gap symmetry and the position of the gap at the Fermi surface.[31]

V. SUMMARY

In CeCoIn$_5$, the $1/T_1$ varies nearly as $T^{1/4}$ in the normal state. In CeIrIn$_5$, $1/T_1$ varies as $T^{1/2}$ in the temperature range between 10 and 80 K, which are well explained as these compounds located close to the AF instability (quantum critical point). In the superconducting state, the Knight shift K deceases in all directions and $1/T_1$ reveals the anisotropic nature of the superconducting energy gap. These results indicate that CeCoIn$_5$ and CeIrIn$_5$ exhibit anisotropic even parity (probably d-wave) superconductivity.

Note added in proof. A detail thermal conductivity measurement of CeCoIn^5 has recently been performed. The measurement shows that the symmetry of the superconductivity is most likely to be $d_{x^2-y^2}$.[32] The ^{115}In NQR measurement of CeIrln^5 has been recently performed by another group.[33] Their results are similar to ours.

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