

## Unique Spin Dynamics and Unconventional Superconductivity in the Layered Heavy Fermion Compound CeIrIn<sub>5</sub>: NQR Evidence

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We report measurements of the <sup>115</sup>In nuclear spin-lattice relaxation rate ( $1/T_1$ ) between  $T = 0.09$  and 100 K in the new heavy fermion (HF) compound CeIrIn<sub>5</sub>. At  $0.4 \leq T \leq 100$  K,  $1/T_1$  is strongly  $T$ -dependent, which indicates that CeIrIn<sub>5</sub> is much more itinerant than known Ce-based HFs. We find that  $1/T_1 T$ , subtracting that for LaIrIn<sub>5</sub>, follows a  $(\frac{1}{T+\theta})^{3/4}$  variation with  $\theta = 8$  K. We argue that this novel feature points to anisotropic, due to a layered crystal structure, spin fluctuations near a magnetic ordering. The bulk superconductivity sets in at 0.40 K below which the coherence peak is absent and  $1/T_1$  follows a  $T^3$  variation, which suggests unconventional superconductivity with line-node gap.

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The emergence of superconductivity near a magnetic instability in cerium (Ce)-based heavy fermion (HF) compounds is one of the most intriguing phenomena in strongly correlated electron systems. Except for CeCu<sub>2</sub>Si<sub>2</sub> which is superconducting at ambient pressure with  $T_c = 0.65$  K [1], the superconductivity emerges near the quantum critical point (QCP) where the magnetic ordering is suppressed by large applied external pressure in CeIn<sub>3</sub> [2], CeCu<sub>2</sub>Ge<sub>2</sub> [3], CePd<sub>2</sub>Si<sub>2</sub> [4], and CeRh<sub>2</sub>Si<sub>2</sub> [5]. In spite of efforts and progress, however, knowledge about this class of superconductors is still limited because of difficult experimental conditions. The recently discovered new family of Ce-based heavy electron systems, CeMIn<sub>5</sub> ( $M = \text{Rh, Ir}$ ) with  $M = \text{Ir}$  being a superconductor already at ambient pressure [6,7], provides new opportunities for studying the nature of the superconductivity in the vicinity of a magnetic instability, the interplay between magnetic excitations and superconductivity, etc. In particular, CeIrIn<sub>5</sub> is suitable for studies using microscopic experimental probes that can be applied more easily at ambient pressure.

CeMIn<sub>5</sub> ( $M = \text{Rh, Ir}$ ) consists of alternating layers of CeIn<sub>3</sub> and MIn<sub>2</sub>. CeRhIn<sub>5</sub> is an antiferromagnet with  $T_N = 3.8$  K but becomes superconducting below  $T_c = 2.1$  K under a moderate pressure of 1.6 GPa [6]. In CeIrIn<sub>5</sub>, the resistivity is already zero at ambient pressure below 1.2 K, but the Meissner effect and the jump in the specific heat are found only at 0.4 K [7]. The electronic specific heat coefficient  $\gamma$  is found to be 750 mJ/mol K<sup>2</sup> [7], which suggests a large mass enhancement. Recent de Haas-van Alphen oscillation in CeIrIn<sub>5</sub> also reveals a cyclotron mass that is  $\sim 20$  times larger than the band mass, consistent with the specific heat result [8].

In this Letter, we report a measurement using local probe, the <sup>115</sup>In nuclear quadrupolar resonance (NQR) study in CeIrIn<sub>5</sub> down to 90 mK, at zero magnetic field. From the temperature ( $T$ ) dependence of the nuclear spin-lattice relaxation rate ( $1/T_1$ ), we find that CeIrIn<sub>5</sub>

is much more itinerant than known Ce compounds such as CeCu<sub>2</sub>Si<sub>2</sub> [9], and show that this compound is located near a magnetic ordering with anisotropic spin fluctuations due to the layered crystal structure. No anomaly was found at 1.2 K in the NQR quantities, but  $1/T_1$  shows an abrupt decrease at 0.40 K below which the NQR intensity also decreases as does the ac susceptibility, confirming a bulk superconductivity below  $T_c = 0.40$  K. The lack of coherence peak in  $1/T_1$  just below  $T_c = 0.40$  K followed by a power-law  $T$  variation,  $1/T_1 \propto T^3$ , indicates that the superconductivity is of unconventional type with an anisotropic gap. Our results show that CeIrIn<sub>5</sub> bears some resemblance to itinerant, quasi-two-dimensional high- $T_c$  copper oxides.

The single crystal of CeIrIn<sub>5</sub> was grown by the In-flux method as in Ref. [6]. X-ray diffraction indicated that the compound is single phase and forms in the primitive tetragonal HoCoGa<sub>5</sub> type structure. The resistivity already drops to zero at 1.2 K, which is in agreement with the reported property [7]. The single crystal was crushed into powder to allow a maximal penetration of oscillating magnetic field,  $H_1$ . The measurements below 1.4 K were performed by using a <sup>3</sup>He/<sup>4</sup>He dilution refrigerator. A small  $H_1$  was used to avoid possible heating by the rf pulse. There are two inequivalent crystallographic sites of In in this compound, In(1) in the CeIn<sub>3</sub> plane and In(2) in the IrIn<sub>2</sub> plane. Two sets of In NQR lines corresponding to these two sites were observed as shown in Fig. 1. The first set of the NQR lines that are equally spaced is characterized by  $\nu_Q = 6.065 \pm 0.01$  MHz and the asymmetry parameter  $\eta = 0$ . The second set of lines that are unequally spaced was found at the positions centered at 33.700, 38.350, 52.185, and 71.432 MHz, respectively, which correspond to  $\nu_Q = 18.175 \pm 0.01$  MHz and  $\eta = 0.462 \pm 0.001$ . Here  $\nu_Q$  and  $\eta$  are defined as  $\nu_Q \equiv \nu_z = \frac{3}{2I(2I-1)\hbar} e^2 Q \frac{\partial V}{\partial z}$ , and  $\eta = |\nu_x - \nu_y|/\nu_z$ , with  $Q$  being the nuclear quadrupolar moment,  $I = 9/2$

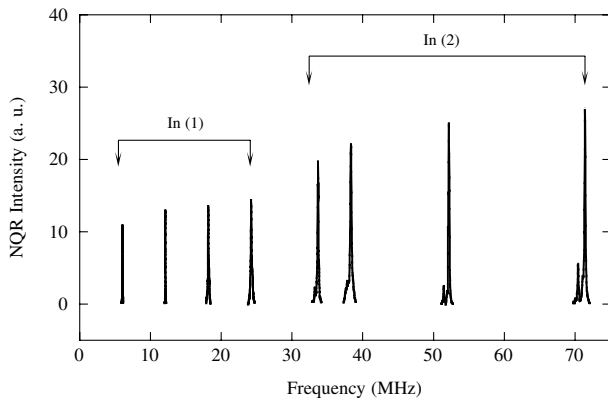


FIG. 1. Two sets of  $^{115}\text{In}$  NQR lines observed at  $T = 4.2$  K corresponding to the In(1) site in the  $\text{CeIn}_3$  plane and the In(2) site in the  $\text{IrIn}_2$  plane. The small peaks to the left of each line of In(2) are from the less abundant isotope  $^{113}\text{In}$ .

being the nuclear spin, and  $\frac{\partial V}{\partial \alpha}(\alpha = x, y, z)$  being the electric field gradient at the position of the nucleus [10]. The symmetric lines are assigned to the In(1) site and the asymmetric lines to the In(2) site, respectively, since the crystallographically In(1) site is axially symmetric but In(2) is not. This assignment is consistent with the observation in  $\text{CeIn}_3$  where  $\eta$  is zero [11].  $\nu_Q$  for In(1) is smaller, but  $\nu_Q$  for In(2) is larger by about 10% than the respective values in  $\text{CeRhIn}_5$  [12]. The transition lines are narrow with the full width at half maximum (FWHM) being  $\sim 50$  kHz, which indicates a good quality of the crystal.  $1/T_1$  measurements were done mostly on the  $1\nu_Q(\pm 1/2 \leftrightarrow \pm 3/2)$  transition at low  $T$  but at the  $4\nu_Q(\pm 7/2 \leftrightarrow \pm 9/2)$  transition at high  $T$  for the In(1) site. The value of  $1/T_1$  was obtained from the recovery of the nuclear magnetization following a single saturation pulse and an excellent fitting to the equations given by MacLaughlin [13]. The value of  $1/T_1$  measured at different transitions shows excellent agreement.

Figure 2 shows  $1/T_1$  as a function of  $T$  in the temperature range of  $0.09 \leq T \leq 100$  K. We discuss the normal-state property above  $T_c$  first. Remarkably,  $1/T_1$  shows strong  $T$  dependence up to the highest temperature that we have measured, 100 K. This is to be compared to other Ce compounds, such as  $\text{CeCu}_2\text{Si}_2$  where  $1/T_1$  becomes  $T$  independent above  $\sim 10$  K [9], which is assigned to be the Kondo temperature,  $T_K$  below which the localized  $4f$  moment is screened to produce a heavy quasiparticle state. This result indicates that  $\text{CeIrIn}_5$  is much more itinerant than other known Ce compounds. In Fig. 3, we plot  $1/T_1 T$  as a function of  $T$ . The inset shows a log-log plot that displays more clearly the behavior just above  $T_c$ . For comparison, we also show the data  $1/T_1 T = 0.81 \text{ sec}^{-1} \text{ K}^{-1}$  for  $\text{LaIrIn}_5$  where no  $4f$  spins are present. The measurement for  $\text{LaIrIn}_5$  was carried out at  $4\nu_Q = 23.77$  MHz. It is seen that  $1/T_1 T$  of  $\text{CeIrIn}_5$  is largely enhanced over that of  $\text{LaIrIn}_5$  and increases strongly with decreasing

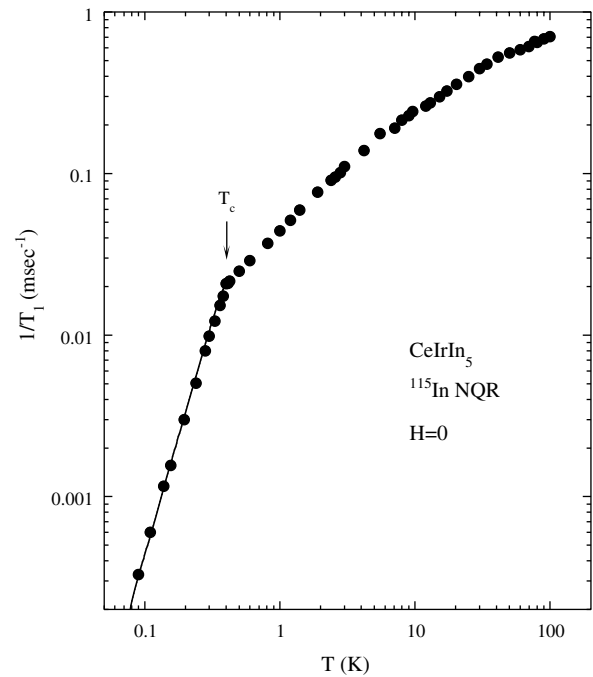


FIG. 2.  $T$  dependence of the  $^{115}\text{In}$  nuclear spin-lattice relaxation rate. The solid curve is a calculation by assuming a line-node gap  $\Delta(\phi) = \Delta_0 \cos \phi$  with  $\Delta_0 = 2.5k_B T_c$  (see text for detail).

$T$ . Also note that a  $T_1 T = \text{const}$  relation, that would be expected from the Landau-Fermi liquid theory, is not obeyed. These aspects indicate that  $1/T_1$  in  $\text{CeIrIn}_5$  is dominated by itinerant spin fluctuations (SFs). In fact, it was found more recently that substituting 40% of Rh for Ir results in an antiferromagnetic (AF) ordering of the system at  $T_N = 2.7$  K [14], which also suggests that  $\text{CeIrIn}_5$  is a nearly AF metal. In Fig. 4 we show  $T_1 T$  as a function of  $T$ . The open circles above 0.4 K correspond to  $1/T_1 T = 1/T_1 T(\text{CeIrIn}_5) - 0.81 \text{ sec}^{-1} \text{ K}^{-1}$  ( $\text{LaIrIn}_5$ ), which represent the contribution due to the presence of Ce  $4f$  spins alone. Note that this correction, by subtracting the relaxation of  $\text{LaIrIn}_5$  which represents other relaxations including the In orbital contribution, is negligibly small for  $0.4 \leq T \leq 15$  K. As seen in the figure, the data can be fitted to a relation of  $T_1 T = C(T + \theta)^{3/4}$  with  $\theta = 8$  K and  $C = 4.75 \text{ msec K}^{1/4}$  (solid curve).

This unique  $T$  dependence of  $1/T_1 T$  has never been observed in other HF compounds. We argue that this novel feature arises from *anisotropic* AF spin fluctuations, due to the *layered* crystal structure of  $\text{CeIrIn}_5$ . In  $d$ -electron, and also  $f$ -electron weak or nearly AF metals, many physical quantities can be explained by the self-consistent renormalization theory for SFs [15]. In this theory, it was shown that the staggered susceptibility at the AF wave vector  $q = Q$ ,  $\chi_Q$  (or the squared magnetic correlation length) follows a Curie-Weiss (CW) variation above the Néel temperature in a weak AF magnet due to mode-mode coupling of SFs, namely,  $\chi_Q \propto \frac{1}{T + \theta}$ . The value  $|\theta|$  is just the Néel temperature ( $T_N$ ) here. In a nearly AF metal

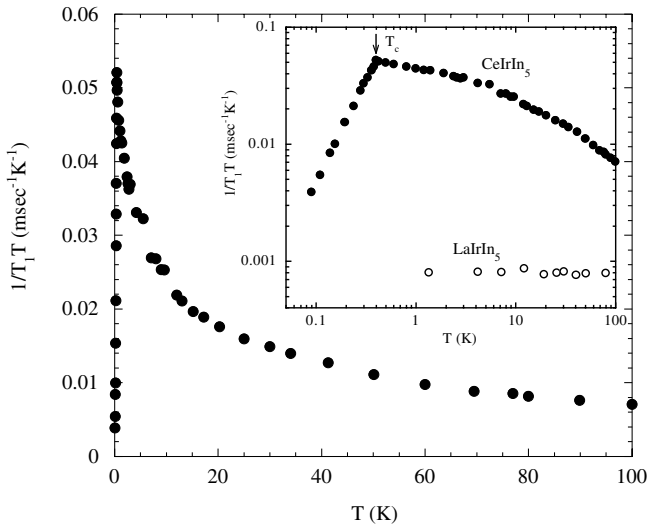


FIG. 3. The  $1/T_1T$  of  $^{115}\text{In}$  plotted as a function of  $T$ . The inset shows a log-log plot that emphasizes the low- $T$  behavior just above  $T_c$ . Also shown, in the inset, are data for  $\text{LaIrIn}_5$ . A strong increase of  $1/T_1T$  with lowering  $T$  points to the antiferromagnetic spin correlation.

that does not order at finite temperature,  $\chi_Q$  is shown to also obey a CW variation, while in this case  $\theta$  measures the closeness of a system to the magnetic ordering;  $\theta$  decreases towards zero upon approaching the ordering. Now, by assuming  $\chi(Q+q, \omega)^{-1} = \chi_Q^{-1} + aq^2 - i\frac{\omega}{T}$  it is shown that  $1/T_1T \propto \chi_Q$  for two-dimensional (2D) systems, but  $1/T_1T \propto \sqrt{\chi_Q}$  for 3D systems [15]. Indeed, in many quasi-2D high- $T_c$  cuprates it is found  $1/T_1T \propto \frac{1}{T+\theta}$  where the value of  $\theta$  decreases upon approaching the mag-

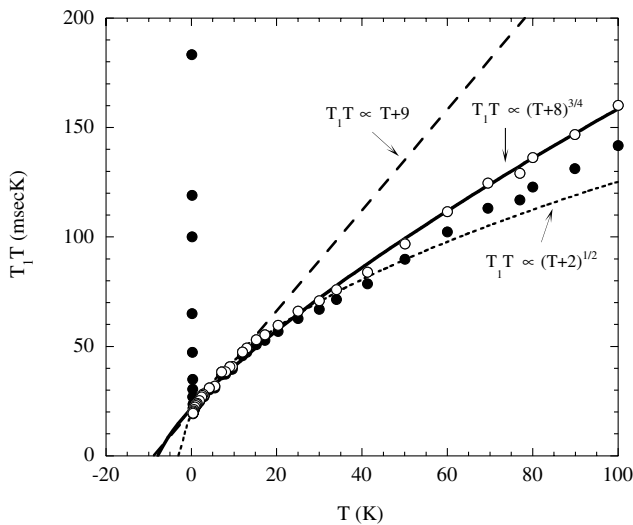


FIG. 4.  $T_1T$  plotted as a function of  $T$ . Solid circles are the raw data of  $\text{CeIrIn}_5$ , while open circles correspond to the normal-state relaxation rate subtracting that of  $\text{LaIrIn}_5$ . The broken line, solid, and dotted curves are the  $T$  variations of  $T_1T \propto T + \theta$  (2D SFs),  $T_1T \propto (T + \theta)^{3/4}$  (anisotropic SFs), and  $T_1T \propto (T + \theta)^{1/2}$  (3D SFs), respectively.

netic ordering. For example, in the so-called overdoped compound  $\text{TlSr}_2\text{CaCu}_2\text{O}_{6.8}$  which is far away from the magnetic ordering,  $\theta$  is 235 K [16]. In a less hole-doped system,  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ,  $\theta = 120$  K for  $x = 0.24$  while it decreases linearly with decreasing hole doping, reaching to  $\theta = 20$  K at  $x = 0.075$  [17]. In AF ordered 3D HF compounds, on the other hand,  $1/T_1T \propto (\frac{1}{T+\theta})^{1/2}$  is well obeyed [18]. The predicted results by the 2D or 3D models are shown in Fig. 4. As can be seen in the figure, although both models capture the low- $T$  behavior, neither of them fits the data in the high- $T$  range. Let us now consider a situation where the dispersion of the SFs is in between 2D and 3D ones. If the SF dispersion in one direction is flat, namely, the magnetic correlation length ( $\xi$ ) is much shorter in one direction than in the other, then by assuming  $\chi(Q+q)^{-1} = \chi_Q^{-1} + a_1(q_x^2 + q_y^2) + a_2q_z^4$  instead of isotropic quadratic dispersion [19], it is shown that  $1/T_1T \propto \chi_Q^{3/4}$ . This anisotropic SF model explained the dynamical susceptibility in  $d$ -electron antiferromagnet  $\text{YMn}_2$ , which orders at  $T_N = 110$  K but the ordering can be suppressed either by applying external pressure or by substituting Sc for Y. In paramagnetic  $\text{Y}_{0.97}\text{Sc}_{0.03}\text{Mn}_2$ , the inelastic neutron scattering measurement found that  $\xi$  is shorter along the  $[001]$  direction ( $\xi_{\perp} = 1.72 \text{ \AA}$ ) than that along the  $[110]$  direction ( $\xi_{\parallel} = 2.86 \text{ \AA}$ ), which is ascribed to the geometrical frustration of the magnetic interaction [20]. Indeed, the same  $T$  variation as found here, namely,  $\frac{1}{T_1T} \propto (\frac{1}{T+\theta})^{3/4}$  was observed in paramagnetic  $\text{YMn}_2$  under pressure [21]. On the above basis, we propose that the  $T$  variation of  $1/T_1T$ , that can be fitted to  $(\frac{1}{T+\theta})^{3/4}$  in the entire  $T$  range except near  $T_c$ , is due to anisotropic spin fluctuations in  $\text{CeIrIn}_5$ . In fact,  $\text{CeIrIn}_5$  has a layered crystal structure. Because of this 2D-like structure, a weaker magnetic correlation along the  $c$  axis can be expected. Further investigation by an inelastic neutron scattering measurement would be interesting to confirm the SF dispersion in this compound. More systematic NQR/NMR study is also underway to see if the deviation of the low- $T$  data from the anisotropic SF curve points to any possible crossover to a different SF regime upon lowering  $T$ . In any case, the small value of  $\theta < 10$  K indicates that  $\text{CeIrIn}_5$  is located in close proximity to the magnetic ordering. Finally, we remark that such strong SFs may also make an appreciable contribution to the huge specific heat.

Next, we discuss the superconducting (SC) state. First, as seen in Fig. 2, no anomaly was detected in  $1/T_1$  around 1.2 K below which resistivity is zero. We also checked carefully the intensity and the linewidth of the NQR spectrum below 1.4 K; no anomaly is found when passing through 1.2 K. However,  $1/T_1$  decreases abruptly at  $T = 0.40$  K, below which the NQR intensity decreases as does the ac susceptibility due to the Meissner effect. These results indicate that the bulk superconductivity sets in at 0.40 K, which is in good agreement with the specific heat measurement [7]. The property of the SC state below

0.40 K is remarkable. Namely,  $1/T_1$  shows no coherence peak just below  $T_c$ , and decreases in proportion to  $\sim T^3$  upon lowering  $T$ . This behavior is not compatible with the isotropic  $s$ -wave gap. Rather, our result is qualitatively similar to that in other HF superconductors, such as  $\text{CeCu}_2\text{Si}_2$  [9],  $\text{UBe}_{13}$  [22], etc. [23,18], and also high- $T_c$  cuprate superconductors [24], which indicates that the SC energy gap is anisotropic. In terms of density of states (DOS),  $T_{1s}$  in the SC state is expressed as  $\frac{T_{1s}}{T_{1s}} = \frac{2}{k_B T} \iint (1 + \frac{\Delta^2}{EE'}) N_s(E) N_s(E') f(E) [1 - f(E')] \delta(E - E') dE dE'$ , where  $N_s(E)$  is the DOS in the SC state,  $f(E)$  is the Fermi function,  $\Delta$  is the energy gap, and  $C = 1 + \frac{\Delta^2}{EE'}$  is called the coherence factor. In an isotropic  $s$ -wave superconductor, the divergence of  $N_s$  at  $E = \Delta$  results in the coherence peak of  $1/T_1$  just below  $T_c$ , and  $1/T_1$  decreases as  $\exp(-\Delta/k_B T)$  at low  $T$  because  $N_s = 0$  for  $E < \Delta$ . In contrast, an anisotropic gap generally reduces the divergence of  $N_s$  and produces a finite DOS at low energy. For example, if we assume a line-node gap of  $\Delta(\phi) = \Delta_0 \cos \phi$ , then by integrating  $C$  and  $N_s = \frac{N_0 E}{\sqrt{E^2 - \Delta(\phi)^2}}$  over  $\phi$  we obtain  $C = 1$ ,  $N_s = \frac{\pi}{2} \frac{N_0 E}{\Delta_0}$  for  $E \leq \Delta_0$ , and  $N_s = \frac{N_0 E}{\Delta_0} \sin^{-1}(\frac{\Delta_0}{E})$  for  $E \geq \Delta_0$ . The finite value of  $N_s$  at  $E = \Delta_0$  removes the coherence peak and the  $E$ -linear DOS below  $\Delta_0$  gives rise to a  $T^3$  variation of  $1/T_1$  at low  $T$ . The curve below  $T_c$  in Fig. 2 depicts the calculated result assuming the above model with  $2\Delta_0 = 5.0k_B T_c$  and a BCS  $T$  dependence for  $\Delta_0$ . This gap amplitude  $\Delta_0$  is about the same as that for  $\text{CeCu}_2\text{Si}_2$  to which should the same model be applied. It is, however, substantially smaller than that in some uranium (U)-based HF superconductors where  $2\Delta_0$  would reach  $\sim 10k_B T_c$  for the same gap function [22,23], which may be related to its proximity to a magnetic instability of the present compound. In fact, a recent study found a reduced  $\Delta_0$  in  $\text{Ce}_{0.99}\text{Cu}_{2.02}\text{Si}_2$  [25], which is believed to be located closer to QCP than a stoichiometric compound [26]. Applying external pressure increases  $\Delta_0$  [25]. In  $\text{CeIrIn}_5$ , applying pressure increases  $T_c$  [7]. Further investigations by NQR under pressure are in progress in order to reveal how the gap amplitude evolves with pressure, and the intimate relation between the superconductivity and the magnetic excitations.

In summary, we find from NQR  $1/T_1$  measurement that the new HF compound  $\text{CeIrIn}_5$  is much more itinerant than known Ce-based HF's. We further find that  $1/T_1 T$ , subtracting that for  $\text{LaIrIn}_5$ , follows a  $(\frac{1}{T+\theta})^{3/4}$  variation with a small  $\theta = 8$  K, and interpret it as arising from anisotropic, due to the layered crystal structure, spin fluctuations near a magnetic ordering. Below  $T_c = 0.40$  K,  $1/T_1$  decreases

in proportion to  $T^3$ , which indicates unconventional superconductivity with an anisotropic energy gap. Our results show that the superconducting state of this new compound is reminiscent of those in known HF and high- $T_c$  cuprate superconductors, but the normal-state magnetic excitation lies in between those for the quasi-2D high- $T_c$  cuprates and the known 3D, more localized HF's. Therefore, the knowledge about this compound may also be useful for understanding the physics of both high- $T_c$  cuprates and traditional HF superconductors.

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