Heavy Fermion Superconductor Ce_2PdIn_8 studied by ^{115}In Nuclear Quadrupole Resonance

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Abstract. We have performed nuclear quadrupole resonance (NQR) measurements on a recently-discovered heavy-fermion superconductor Ce_2PdIn_8 with superconducting critical temperature $T_c = 0.64$ K. Below coherent temperature $T_{coh} \sim 30$ K, the spin-lattice relaxation rate $1/T_1$ decreases with decreasing temperature T and is proportional to $T^{1/2}$ between T_c and T_{coh}. This is clearly different from the Fermi-liquid behavior in which the T dependence is proportional to T, and indicates that Ce_2PdIn_8 is located on the verge of antiferromagnetic quantum critical point from the view point of the NQR. Below T_c, $1/T_1$ shows no coherence peak and is proportional to T^3. This is clear evidence for the realization of unconventional superconductivity with line nodes in this compound.

1. Introduction

The relationship between superconductivity and magnetic spin fluctuations is one of the central issues in strongly correlated electron systems [1]. Unconventional superconductivity has been discovered in pressurized $CeIn_3$ [2], pressurized $CeRhIn_5$ [3], and $CeCoIn_5$ [4] near the magnetic instability point. In these systems, magnetic correlation arises from the competition and/or cooperation between the Ruderman-Kittel-Kasuya-Yosida interaction and the Kondo effect which are based on the hybridization effect of localized f-electrons and itinerant conduction electrons. It has been recognized that the dimensionality of a system also plays a vital role in unconventional superconductivity near a quantum critical point (QCP) [5]. Such importance has recently been recognized by experimentally investigating artificial superlattices of the antiferromagnetic (AF) heavy-fermion (HF) compound $CeIn_3$ and the conventional metal $LaIn_3$ [6]. Ce-based HF compounds with the formula Ce_nMIn_{3n+2} ($n = 1, 2, \infty$, $M =$ Co, Rh, Pd, Ir) are also good candidates for the investigation of dimensionality, quantum criticality, and superconductivity [2, 3, 4, 7, 8, 9, 10, 11, 12], because their crystal structure can be viewed as successive layers of quasi-two-dimensional $CeMIn_5$ and/or three-dimensional $CeIn_3$.

Among the quasi-2D HF superconductors, $CeCoIn_5$ is one of the most intensively studied systems. This compound clearly exhibits non-Fermi-liquid (NFL) behavior in the normal state [4] and d-wave superconductivity below the superconducting (SC) transition temperature $T_c = 2.3$ K [10]. The outstanding phenomenon of $CeCoIn_5$ is a magnetic field-induced spin-density-wave state (so-called Q phase) that forms in the low-temperature and high-field (LTHF) corner of the $H - T$ phase diagram [13, 14]. Most importantly, it has been suggested that this

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new phase may be the long sought inhomogeneous Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) SC state [15].

Recently superconductivity was discovered in Ce_2PdIn_8 with $T_c \simeq 0.7$ K [9]. This compound has a Ho_2CoGa_8-type tetragonal crystal structure (space group $P4/mmm$). The electronic specific heat coefficient is approximately 1.5 J/K^{2}mol-fu [9, 16, 17, 18], which indicates heavy-fermion superconductivity. Resistivity has clearly shown a logarithmic dependence on the temperature T between 40 and 100 K with a local maximum at approximately 30 K($\sim T_{\rm coh}$) corresponding to a cross-over temperature between the coherent and incoherent dense Kondo regimes and a linear dependence on T between T_c and approximately 10 K, which is evidence for NFL behavior [9, 16, 17]. Moreover, it has been clarified recently that the magnetic field H and/or pressure can tune the QCP [18, 19, 20, 21, 22]. As for the superconductivity, it has been shown that the most probable candidate for the SC gap symmetry is d-wave superconductivity. Thermal conductivity measurements under magnetic field have clearly shown the presence of residual density of states (R-DOS) at the Fermi level [20]. Penetration depth measurements using a tunnel diode resonator have also indicated a nodal-line structure in the SC gap [23]. All these properties found in Ce_2PdIn_8 are quite similar to those in the more two-dimensional $CeCoIn_5$. However, the T_c and upper critical field H_{c2} under zero and finite pressure for Ce_2PdIn_8 are considerably lower than those for $CeCoIn_5$. This is probably associated with the dimensionality of the systems.

These fascinating phenomena in Ce_2PdIn_8 have been revealed by bulk-property measurements such as electrical transport measurements. Therefore, it is vital to perform microscopic and site-selective measurements to clarify the magnetic properties in the normal state and the superconductivity of this compound. In this paper, we report ^{115}In nuclear quadrupole resonance (NQR) measurements of Ce_2PdIn_8.

2. Experiments

Polycrystalline samples of Ce_2PdIn_8 were synthesized by the arc melting method. The detailed procedure is described in Ref. [17]. The ^{115}In NQR measurements were performed using a phase-coherent pulsed NQR spectrometer in the frequency range of 7-60 MHz. The crystals were powdered in order to reduce heating up of the samples at low temperature and improve signal intensity. Measurements above 1.5 K were performed using a ^{4}He cryostat and below 1.5 K with a ^{3}He-^{4}He dilution refrigerator. The spin-lattice relaxation time T_1 was obtained from the recovery of the nuclear magnetization after a saturation pulse. T_c was estimated to be 0.64 K through ac susceptibility by measuring the temperature dependence of the characteristic frequency of the NQR sample-coil $f_{\rm coil}$.

3. Results and Discussion

There are three inequivalent crystalline In sites in Ce_2PdIn_8: the most symmetric site In(1) ($4mm$), the less symmetric site In(2) ($mmm.$), and the least symmetric site In(3) ($2mm.$). By diagonalizing the electric quadrupole $\mathcal{H}_Q$ for the In nuclei ($I = 9/2$) and considering the three inequivalent In sites, we can assign all the In sites in the NQR spectrum of Ce_2PdIn_8. All the feature of the NQR spectrum is shown elsewhere [24]. For each In site, we denote the resonance lines $1\nu_Q$, $2\nu_Q$, $3\nu_Q$ and $4\nu_Q$ according to increasing frequency. The $1\nu_Q$ line is from the transition between $|I_z = \pm 1/2\rangle$ and $|I_z = \pm 3/2\rangle$; the $2\nu_Q$ line is from the transition between $|I_z = \pm 3/2\rangle$ and $|I_z = \pm 5/2\rangle$, etc.

As clearly shown in Fig. 1, we did not observe any signals other than those from the ^{113}In and ^{115}In nuclei from Ce_2PdIn_8. Because natural abundance of ^{113}In is 4.3% and that of ^{115}In is 95.7%, the ^{113}In line intensities are quite small. The lines due to ^{113}In overlap with those of ^{115}In because the ratio of quadrupole moments is $^{113}Q/^{115}Q = 0.82/0.83$. This confirms the quality of the samples that were free of any clear parasitic phases which were always found

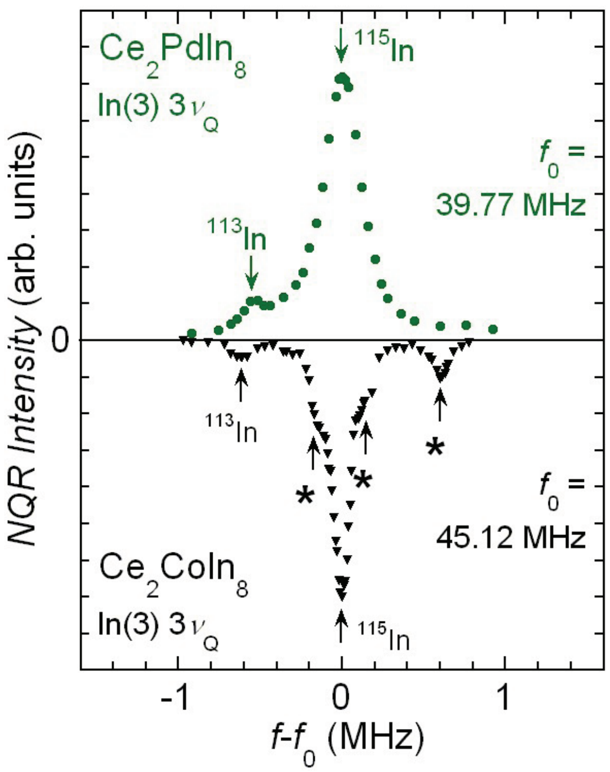

Figure 1. (Color online) In-NQR spectra of Ce_2PdIn_8 and Ce_2CoIn_8. The data of Ce_2CoIn_8 was cited from ref. [12].

in Ce_2CoIn_8 (* marks in Fig. 1) and Ce_2RhIn_8 [12]. The line-width is about 1.8 times wider than the corresponding line-widths of single-crystalline $CeCoIn_5$ [10] and Ce_2CoIn_8 [12]. This line-width broadening may be due to some structural disorder in the sample studied or/and may arise from the averaged electric-field-gradient in the powdered samples.

We obtained T_1 at the frequency of the spectral center at each In site over the range of 0.075-70 K. The nuclear magnetization recovery curve was fitted by the quadruple exponential function as expected for each line of the spectrum of the nuclear spin $I = 9/2$ of the ^{115}In nuclei [25]. For In sites with finite asymmetry parameter η, we adopted the modification of the recovery curves given by Chepin and Ross [26]. All the data were well fitted to the corresponding curve with a single T_1 component even at the lowest measurement temperature.

Obtained temperature dependence of $1/T_1$ is displayed elsewhere [24]. Instead, we show temperature dependence of the spin-lattice relaxation rate $1/T_1$ at the In(1) site of Ce_2PdIn_8, $CeCoIn_5$, and $CeIrIn_5$, normalized at the respective T_c in Fig. 2. In the normal state of Ce_2PdIn_8, the temperature dependence of $1/T_1$ remains constant at higher temperatures above about 30 K($\sim T_{coh}$). This indicates that the $4f$ electrons of the Ce ions are nearly localized above this temperature. This is consistent with the behavior that the resistivity possesses a broad peak around T_{coh} and exhibits a $-\log T$ temperature dependence between 40 and 100 K [9, 16, 17]. $1/T_1$ varies nearly as $T^{1/2}$ between 0.7($\simeq T_c$) and 30 K. This is clearly different from the Fermi-liquid behavior in which the T dependence is proportional to T (namely, the Korringa relation) and is consistent with the NFL behavior confirmed in various previous measurements [9, 16, 17, 18, 19, 20, 21, 22]. However, in order to discuss the difference of the normal state properties of these compounds shown here, we need Knight shift measurements related with transferred hyperfine field from Ce moments and more detailed data at higher

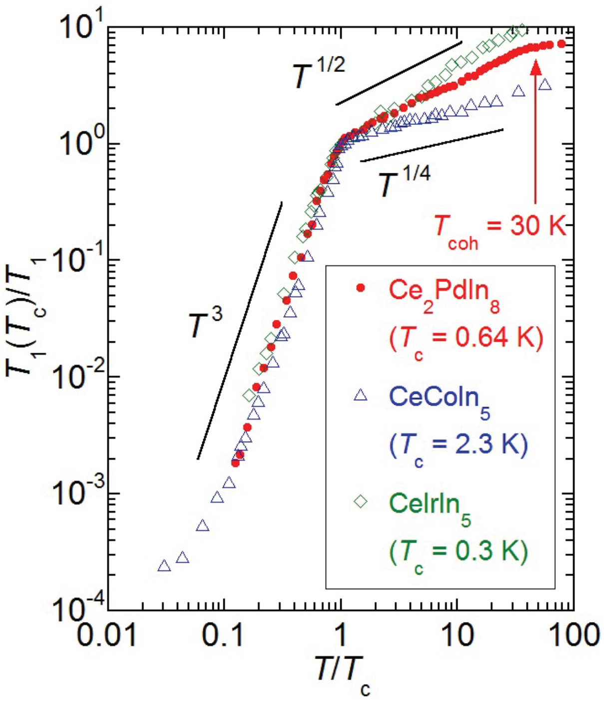

Figure 2. (Color online) Temperature dependence of the spin-lattice relaxation rate $1/T_1$ at the In(1) site of Ce_2PdIn_8, $CeCoIn_5$ and $CeIrIn_5$, normalized at the respective T_c. The $1/T_1$ data of $CeCoIn_5$ and $CeIrIn_5$ were taken from Ref. [10].

temperature above approximately 100 K related with contribution from localized f electrons and conduction electrons.

In the SC state, $1/T_1$ decreases rapidly just below T_c and the Hebel-Slichter coherence peak was not observed. $1/T_1$ varies nearly proportional to T^3 at low temperatures. We did not observe any saturation or T linear dependence of $1/T_1$ at lowest measurement temperatures, which would arise from residual density of states often caused by a small amount of paramagnetic impurities in nodal superconductors. This is because the present lowest measurement temperature of 75 mK is approximately $T_c/9$ and the temperature is not sufficiently low to observe such an extrinsic contribution. As apparent from Fig. 2, there is a good agreement of temperature dependence of Ce_2PdIn_8, $CeCoIn_5$ and $CeIrIn_5$ in the SC state. This coincidence is attributable to the similar energy gap amplitudes in these compounds. The similar gap sizes for both compounds suggest that the SC states of Ce_2PdIn_8 and $CeCoIn_5$ are quite similar to each other. Hence, we may speculate that the SC symmetry of this compound is *d*-wave. However, in order to conclude on this issue, we must clarify the SC symmetry of the spin part of this compound by measuring the Knight shift with a single crystal.

4. Summary

We have performed NQR measurements on the HF superconductor Ce_2PdIn_8 with $T_c = 0.64$ K. Below $T_{coh} \sim 30$ K, $1/T_1$ decreases with decreasing temperature and varies nearly as $T^{1/2}$ between T_c and T_{coh}. This is clearly different from the Fermi-liquid behavior in which the T dependence is proportional to T, and indicates that Ce_2PdIn_8 is located close to the AF QCP from the viewpoint of NQR. Below T_c, $1/T_1$ shows no coherence peak and is proportional to T^3. This is clear evidence of unconventional superconductivity with line nodes. Further studies under finite magnetic field and/or pressure are also needed to understand the properties of the field-/pressure-induced QCP and the LTHF SC phase of the compound.

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