

In-NQR study of heavy fermion superconductor Ce₂PdIn₈ under pressure

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Abstract. ¹¹⁵In nuclear quadrupole resonance measurements were performed in the normal state of the heavy fermion superconductor Ce₂PdIn₈ under hydrostatic pressure up to about 2.3 GPa. The observed behavior of the spin-lattice relaxation rate revealed a systematic suppression of antiferromagnetic critical fluctuations with increasing pressure.

1. Introduction

It has been theoretically recognized that dimensionality plays a key role in unconventional superconductivity (SC) emerging near a quantum critical point (QCP) [1], and Ce-based heavy-fermion intermetallics with the general formula Ce_nTIn_{3n+2} ($n = 1, 2, \infty$, $T = \text{Co, Rh, Ir, Pd, Pt}$) have been revealed good candidates for systematic experimental study of this problem [2]. The quasi-two-dimensional (2D) compounds CeCoIn₅ and CeRhIn₅ are superconductors with $T_c = 2.3$ K (0 GPa) [3] and $T_c = 2.2$ K (2.5 GPa) [4], respectively, while the most 3D phase CeIn₃ becomes superconducting under pressure of $P \simeq 2.7$ GPa with $T_c = 0.2$ K [5]. These findings are fully in line with the expectation that the critical temperature in spin-fluctuation-driven superconductors decreases with increasing the system dimensionality.

The compounds Ce₂TIn₈ crystallizing with the crystal structure of the Ho₂CoGa₈ type are believed to have a character intermediate between 2D- and 3D-like [2, 6]. Recently, ambient pressure SC was discovered in Ce₂PdIn₈ with $T_c \simeq 0.7$ K [7]. The electronic specific heat coefficient is approximately 1.5 J/K²mol-fu [7, 8, 9, 10], which implies a heavy-fermion ground state. The electrical resistivity of the compound exhibits a logarithmic temperature dependence above 40 K, with a local maximum at approximately 30 K ($\sim T_{\text{coh}}$), marking a crossover between the coherent and incoherent dense Kondo regimes, and a linear temperature dependence below about 10 K down to T_c , evidencing a non-Fermi-liquid (NFL) behavior near QCP [7, 8, 9]. Moreover, it has been clarified that QCP can be tuned by magnetic field and pressure [10, 11, 12, 13, 14]. As for the superconductivity, it has been shown that d -wave is the most probable candidate for the SC gap symmetry. The thermal conductivity data have revealed the presence of residual density of states (R-DOS) at the Fermi level [12]. Penetration depth measurements [15] and nuclear quadrupole resonance (NQR) studies [16] have also indicated a nodal-line structure in the superconducting gap. The SC properties of Ce₂PdIn₈ are thus quite similar to those reported for CeCoIn₅. However, T_c and the upper critical field $H_{c2}(0)$ of



Ce_2PdIn_8 are considerably lower than the values found for CeCoIn_5 . The difference is probably associated with the fact that the crystal structure of the latter system is much more two-dimensional than that of Ce_2PdIn_8 .

The high-pressure electrical resistivity measurements have revealed that both the superconductivity and the NFL character in Ce_2PdIn_8 are gradually suppressed with increasing pressure. To shed more light on the quantum critical behavior of the compound, it is vital to perform microscopic and site-selective experiments under pressure. In this paper, we report on our high-pressure ^{115}In NQR studies on the normal state of Ce_2PdIn_8 , and compare the results with the bulk property data.

2. Experiments

A polycrystalline sample of Ce_2PdIn_8 was synthesized by arc melting and characterized for its quality, as described in Ref. [9]. The ^{115}In NQR studies were performed in the frequency range of 7–60 MHz using a phase-coherent pulsed NQR spectrometer. Measurements were carried out above 1.5 K using a ^4He cryostat. The spin-lattice relaxation time T_1 was obtained from the recovery of the nuclear magnetization after a saturation pulse. The high pressure was generated using a BeCu/NiCrAl piston-cylinder cell filled with Daphne 7373 oil. The NQR frequency of the ^{63}Cu resonance in Cu_2O at 4.2 K was used as an indicator of the pressure value inside the cell [17, 18].

3. Results and Discussion

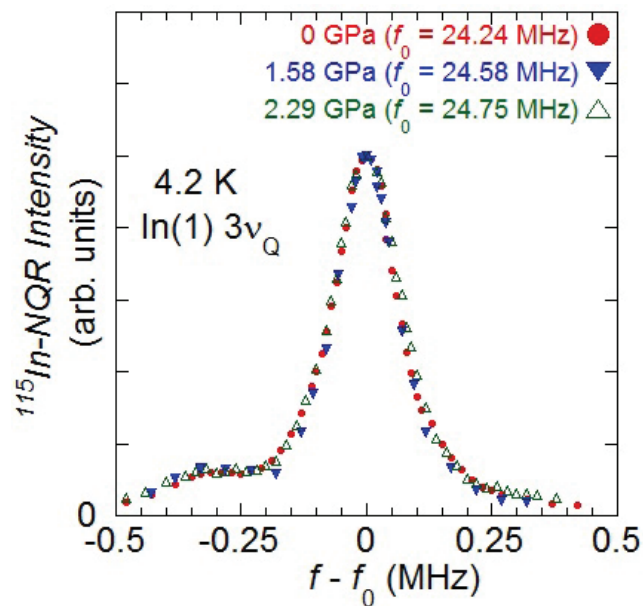


Figure 1. (Color online) In-NQR spectra of Ce_2PdIn_8 at the In(1) site for the $3\nu_Q$ line (the transition between $|I_z = \pm 5/2\rangle$ and $|I_z = \pm 7/2\rangle$). For the sake of comparison, the spectra obtained at different pressures were shifted to 0 MHz.

Figure 1 shows the typical NQR spectra of Ce_2PdIn_8 taken at 4.2 K under pressures of 0, 1.58, 2.29 GPa (the site index and the NQR line number are the same as those defined in Ref. [16]). Narrow NQR lines (~ 160 kHz) and their nearly identical shape indicate high quality of the sample and good hydrostatic pressure homogeneity inside the cell. A broad shoulder around $f - f_0 \sim 0.3$ MHz is originating from the signal of ^{113}In nuclei as previously reported at ambient pressure in Ref. [16]. This also confirms the quality of the specimen. The frequency position f_0 of the NQR line center at a given temperature increases with increasing pressure (the same trend was observed for all the other NQR lines measured), and this feature can be ascribed to the compression of the unit cell volume.

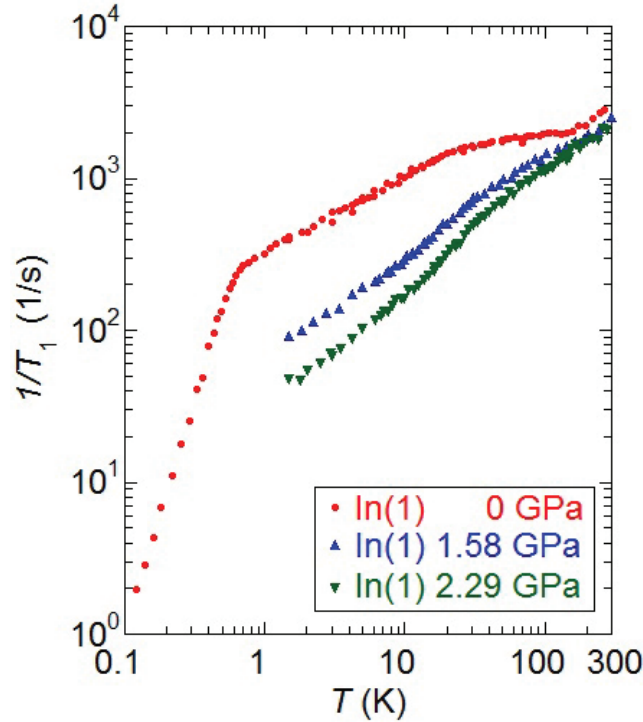


Figure 2. (Color online) Temperature dependence of the spin-lattice relaxation rate $1/T_1$ at the In(1) site of Ce_2PdIn_8 under pressure of 0, 1.58, 2.29 GPa. The ambient pressure data below 1.5 K were taken from Ref. [16].

At ambient pressure, the spin-lattice relaxation time T_1 in Ce_2PdIn_8 was derived over the temperature range 0.075–300 K at the frequency f_0 of the spectral center at the In(1) site. The nuclear magnetization recovery curves were analyzed in terms of the quadruple exponential function, appropriate for the nuclear spin $I = 9/2$ of the ^{115}In nuclei [19]. For example, for the $4\nu_Q$ line with the asymmetry parameter $\eta = 0$ the following formula was applied

$$1 - \frac{m(t)}{m_0} = 0.1212 \exp\left(-\frac{3t}{T_1}\right) + 0.5594 \exp\left(-\frac{10t}{T_1}\right) + 0.297 \exp\left(-\frac{21t}{T_1}\right) + 0.0224 \exp\left(-\frac{36t}{T_1}\right), \quad (1)$$

where $m(t)$ and m_0 denote the nuclear magnetization after a time t from the NMR saturation pulse and the thermal equilibrium magnetization, respectively.

Figure 2 shows the temperature dependencies of the spin-lattice relaxation rate $1/T_1$ in Ce_2PdIn_8 measured under pressures of 0, 1.58, 2.29 GPa. $1/T_1$ was evaluated from the $2\nu_Q$, $3\nu_Q$ and $4\nu_Q$ lines. The results were found to perfectly coincide at 4.2 K for the $2\nu_Q$ and $3\nu_Q$ lines and at 100 K for the $3\nu_Q$ and $4\nu_Q$ lines. It is worth noting that the new data derived at ambient pressure at temperatures above 1.5 K were found in perfect agreement with the previous results obtained on a different sample, reported in Ref. [16]. This finding proves the reliability of our determination of $1/T_1$ for Ce_2PdIn_8 .

At ambient pressure, $1/T_1$ nearly saturates between $T_{\text{coh}} (\simeq 30 \text{ K})$ and about 150 K. This behavior indicates that the $4f$ electrons of the Ce ions are nearly localized above T_{coh} . Actually, above 60 K, one can approximate the experimental data by the function

$$\frac{1}{T_1} = \alpha T + \beta, \quad (2)$$

where the first term corresponds to the Korringa contribution due to correlation-free conduction electrons and the second term accounts for the contribution due to localized $4f$ electrons. The fit parameters are $\alpha = 4.3$ 1/sK and $\beta = 1.5 \times 10^3$ 1/s. It is note worthy that the picture of localized $4f$ electrons at higher temperatures is valid because the value β is dominant in the fitting. The general property of heavy fermion compounds is that the correlation-free conduction electron term is much smaller than the term due to the conduction electrons hybridized at low temperatures with the $4f$ electrons $(1/T_1)_f$. Indeed, in the case of Ce_2PdIn_8 , e.g. at 10 K, $\alpha T = 43$ 1/s is negligibly small in relation to the measured value $1/T_1 = 1000$ 1/s. Therefore, one can estimate the relaxation rate originating from the coherent $4f$ electrons at low temperatures $(1/T_1)_f$ by subtracting the Korringa term from the $1/T_1$ data obtained below T_{coh}

$$\left(\frac{1}{T_1}\right)_f = \frac{1}{T_1} - \alpha T. \quad (3)$$

Assuming that αT is independent of pressure [20], one obtains from the above formula $(1/T_1)_f$ that is close to the raw $1/T_1$ values, yet different enough to discuss the electronic state in Ce_2PdIn_8 more precisely. At a given temperature, the magnitude of $(1/T_1)_f$ systematically decreases with increasing pressure. In the range 1.5-20 K, the slope of $(1/T_1)_f$ versus T becomes steeper with increasing pressure. Altogether, these features indicate a gradual suppression of the antiferromagnetic fluctuations with rising pressure. In order to describe this effect quantitatively one can use the self consistent renormalization (SCR) theory [21]. Within the SCR approach, depending on the effective dimensionality of the systems, the following relationships should be observed

$$\left(\frac{1}{T_1 T}\right)_f \propto \chi_Q(T) \quad (2D \text{ system}), \quad (4)$$

$$\left(\frac{1}{T_1 T}\right)_f \propto \sqrt{\chi_Q(T)} \quad (3D \text{ system}), \quad (5)$$

where the staggered susceptibility $\chi_Q(T)$ with the propagation vector Q follows the Curie-Weiss law

$$\chi_Q(T) \propto \frac{1}{T + \theta}. \quad (6)$$

with a positive Curie-Weiss temperature θ .

Applying the SCR theory to the NQR data of Ce_2PdIn_8 in the range between 1.5 and 15 K one obtains the values of θ collected in Table 1. For both models, the Curie-Weiss temperature increases with increasing pressure. Taking into account that θ is a measure of the distance from the quantum critical point [21], one can conclude that hydrostatic pressure systematically drives the compound away from its QCP. However, one should notice that up to 2.29 GPa, the value of θ remains small, which means that the spin fluctuations keep significant even at that high pressure. These findings are fully consistent with the main result of the electrical resistivity measurements under pressure [13].

As can be inferred from the θ uncertainty values given in Table 1, a better fit to the NQR experimental data of Ce_2PdIn_8 was achieved within the 3D SCR model. This result is at odds with the analyzes of the low-temperature heat capacity data, which led to the preference of the 2D scenario [10]. It seems reasonable to expect that the character of spin fluctuations in Ce_2PdIn_8 is neither as much 2D as found for CeCoIn_5 nor as 3D as evidenced for UIn_3 . In an intermediate situation, bulk property techniques and microscopic probes may appear more sensitive to somewhat different aspects of the dimensionality, and thus lead to seemingly contradictory results.

Table 1. SCR results for the Curie-Weiss temperature of Ce_2PdIn_8 derived within 2D and 3D AF models.

Pressure (GPa)	0	1.58	2.29
θ (K): 2D model	3.3(2)	7.9(4)	11.3(7)
θ (K): 3D model	0.0(1)	1.0(2)	2.3(3)

4. Summary

The heavy fermion superconductor Ce_2PdIn_8 was studied by means of In-NQR measurements under pressure up to about 2.3 GPa. The results indicated a gradual suppression of the antiferromagnetic spin fluctuations with increasing pressure, which manifests a gradual shift of the system away from the quantum critical magnetic instability. The terminal pressure reached in our experiments was however too low to stabilize the Fermi liquid state, hence further NQR investigations of Ce_2PdIn_8 at much higher pressures are needed.

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