Search for the field-induced magnetic instability around the upper critical field of superconductivity in $H \parallel c$ in CeCoIn$_5$

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We present nuclear spin-lattice relaxation rate ($1/T_1$) at the Co site and ac-susceptibility results in the normal and superconducting (SC) states of CeCoIn$_5$ for $H \parallel c$ near the SC upper critical field $H_{c2}$ above 0.1 K. At 4.2 T, $1/T_1$ rapidly decreases below the SC transition temperature, consistent with the previous reports. Although the field dependence of $1/T_1$ at 0.1 K shows a peak at 5.2 T above $H_{c2}$, the temperature dependence of $1/T_1$ at 5.2 T is independent of temperature below 0.2 K, showing a Fermi-liquid behavior. In addition, we found no NMR-spectrum broadening by the appearance of internal fields around $H_{c2}$ at 0.1 K. We could not detect any field-induced magnetic instability around $H_{c2}$ down to 0.1 K although the remarkable non-Fermi-liquid behavior towards $H_{c2}$ was observed in various physical quantities.

KEYWORDS: field-induced criticality, NMR, ac susceptibility, heavy fermion

1. Introduction

Heavy fermion systems have many examples of a quantum critical point (QCP), where magnetic transition temperature is suppressed to zero by applied a pressure or magnetic field, and unconventional superconductivity has been discovered in many cases. This fact provides strong evidence for magnetically mediated Cooper pairing in these superconductors [1–3]. In particular, CeTIn$_5$ ($T =$ Co, Ir, Rh) series, called 1-1-5 family, have attracted much attention for QCP studies because of an emergence of various phenomena originating from 4$f$ electrons such as antiferromagnetic (AFM) order, heavy fermion (FL) state, and superconducting (SC) state under the pressure or magnetic field [4–6].

In this paper, we focus on CeCoIn$_5$. This compound is located in the vicinity of the AFM phase and shows superconductivity at a transition temperature $T_c \approx 2.3$ K at zero-field and ambient pressure [7]. The Knight shift decreased rapidly on entering the SC phase for $H \parallel c$, indicating a singlet Cooper pair forming in CeCoIn$_5$ [8,9]. Nuclear spin-lattice relaxation rate ($1/T_1$) was proportional to $\sim T^3$ far below $T_c$, suggesting that CeCoIn$_5$ is a line-nodal superconductor [9]. In addition, the $d_{x^2-y^2}$ symmetry was suggested from the several measurements such as the field-angle resolved thermal conductivity [10] and heat-capacity [11] measurements and the spectroscopic imaging scanning tunneling microscopy [12, 13] measurement.

For $H \parallel ab$, coexistence of Flude-Ferrell-Larkin-Ovchinnikov (FFLO) state and incommensurate spin-density-wave order (called Q-phase) was observed near $H_{c2}$ [14, 15]. The NMR spectrum at the In(2) site abruptly shifts at $T_c$ with the first order character determined with specific-heat and magnetization measurements near $H_{c2}$, and splits in the Q-phase inside the SC state [15–18]. It is noted that the magnetic Q-phase can exist inside the SC state. On the other hand, the presence of pure
FFLO state in $H \parallel c$ was suggested from the various measurements near $H_{c2}$ [18, 19], but the crucial evidence of the FFLO state in $H \parallel c$ has not been obtained. In addition, the presence of the field-induced magnetic instability similar to the Q phase is another interesting issue for $H \parallel c$ near $H_{c2}$, since a remarkable non Fermi liquid behavior towards $H_{c2}$ was observed in various measurements. Sakai et al. reported that $1/T_1 T$ measured at 5 T increased down to 0.2 K with decreasing temperature due to the development of the quasi-2D spin fluctuations [5], but $1/T_1 T$ below 0.2 K was constant (Korringa-law), indicating the FL state. This was consistent with the dHvA [21], resistibility [22], and thermal conductivity measurements [23]. In the NMR study by Sakai et al., the presence of the maximum of $1/T_1 T$ near $H_{c2}$ was shown and suggested the possibility of the coincidence with the field-induced magnetic critical point and $H_{c2}$ in $H \parallel c$, but the detailed search of the magnetic phase has not been performed [5]. In this paper, we report the detailed temperature and field dependence of $^{59}$Co-NMR and ac susceptibility results for $H \parallel c$ near $H_{c2}$. No superconductivity was observed from the ac susceptibility measurement above 5.0 T and 0.2 K, but $1/T_1 T$ at 0.1 K shows a maximum at 5.2 T above $H_{c2}$. However, $1/T_1 T$ measured at 5.2 T stayed constant below 0.2 K, indicative of the absence of field-induced magnetic instability around $H_{c2}$.

2. Experimental Procedures

Single crystals of CeCoIn$_5$ were synthesized by the In-flux method [20]. We selected the thin-plate single crystal (1.5 mm × 2.0 mm × 0.2 mm) with the $c$ axis normal to the plate, and performed the $^{59}$Co-NMR measurement on the single crystal. The $^{59}$Co-NMR spectra were obtained by summing the Fourier transform spectra from the spin-echo signal obtained at equally spaced rf frequencies at a fixed magnetic field. Since $^{59}$Co nuclear spin is $7/2$ ($I = 7/2$), the $^{59}$Co-NMR spectrum consists of seven peaks. The sharpness of the resonance line indicates the high quality of the crystal. $1/T_1$ at the Co site was obtained from a central line arising from the $|{-1/2}⟩ ⇐⇒ |{+1/2}⟩$ transition. The crystal alignment with respect to the magnetic field was precisely performed with a single-axis rotator in the horizontal field generated by a split-magnet, and the misalignment is within 0.5°.

3. Results

Figure 1(a) shows the crystal structure of CeCoIn$_5$. The crystal structure of CeCoIn$_5$ is the tetragonal HoCoGa$_5$ type structure and the space group $P4/mmm$ [7]. The Ce and Co sites, occupying the 1a and 1b Wyckoff positions with the both point symmetries $4/mmm$, are at the same fourfold axis along the $c$ axis. There are two crystallographically inequivalent In sites, In(1) and In(2), occupying the 1c and 4i Wyckoff positions with the point symmetries $4/mmm$ and $2mm$, respectively.

Figure 1(b) shows the temperature dependence of ac susceptibility $\chi_{ac}$ for various magnetic fields parallel to the $c$ axis. Below 4.8 T, $\chi_{ac}$ decreases below $T_c(H)$, caused by the diamagnetic shielding. On the other hand, above 5.0 T, a clear anomaly is not observed down to 0.2 K, which is consistent with the previous reports ($\mu_0 H_{c2} = 5.0$ T) [7].

Figure 2(a) shows the field dependence of $1/T_1 T$ and $\chi_{ac}$ at 0.1 K near $H_{c2}$ for $H \parallel c$. At 4.8 T, the $^{59}$Co-NMR spectrum was splitting below $T_c$ due to the first order transition, and we follow the normal-state component. $1/T_1 T$ showed a peak at 5.2 T, suggesting that the field-induced peak of $1/T_1 T$ is above $H_{c2}$. Figure 2(b) shows the temperature dependence of $1/T_1 T$ in various magnetic fields parallel to the $c$ axis. Above $T_c$, $1/T_1 T$ increases with decreasing temperature due to the development of AFM fluctuations. In smaller fields than $H_{c2}$ (2.0 and 4.2 T), $1/T_1 T$ abruptly decreases without a coherent peak just below $T_c$, supporting the $d$-wave superconductivity. At 5.2 T, $1/T_1 T$ is independent of temperature below 0.2 K, showing the Korringa behavior even at the critical field where $1/T_1 T$ shows a maximum. This indicates that a field-induced magnetic instability is absent in CeCoIn$_5$ near $H_{c2}$ in $H \parallel c$. 

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Figure 3 (a) shows the $^{59}$Co-NMR spectra measured at 0.1 K in 5.0, 5.2, and 5.4 T near $H_{c2}$. The shape of lines are insensitive to the field, and any linewidth broadening of the $^{59}$Co-NMR spectra was not observed at 5.2 T, as shown the Fig. 3(b), also indicative of the absence of due to the appearance of static internal field a field-induced magnetic instability around $H_{c2}$ as well as the result of $1/T_1$.

Many anomalous states due to the development of spin-fluctuations, such as a non-Fermi liquid behavior, have been observed near QCPs in various heavy-fermion compounds. A typical example is YbRh$_2$Si$_2$, in which $1/T_1$ of $^{29}$Si in YbRh$_2$Si$_2$ continues to increase at the field-induced critical point with decreasing temperature [24]. However, our result shows that $1/T_1$ is constant below 0.2 K, indicating that the FL state holds even at the critical field where $1/T_1$ shows the maximum. The $1/T_1$ behavior observed in the critical region of CeCoIn$_5$ is reminiscent of the criticality observed in CeRu$_2$Si$_2$ showing the meta-magnetic criticality. Sakakibara et al. reported that the FL state holds at low temperatures even at the critical region, and suggested that the origin of this criticality is related with the change of the $f$ electron character from itinerant to localized [25]. $1/T_1$ of CeRu$_2$Si$_2$ at the critical field increased down to $T_M$ and stayed constant below $T_M$ [26], as if CeRu$_2$Si$_2$ possesses a finite crossover temperature at $T_M$ [25]. We suggest that the critical behavior observed in CeCoIn$_5$ near $H_{c2}$ might originate from the field continuous change of the Fermi-surfaces, in which the large Fermi-surfaces still remain above $H_{c2}$ as shown in [21]. The $1/T_1$ behaviors against $H$ and $T$ show the crossover from the non-FL to FL occurring at around 5.2 T below 0.2 K, in good agreement with the recent thermal expansion measurement [27], but inconsistent with the thermal conductivity results showing the non-FL behaviors above $H_{c2}$ [23].

Recently, the field-induced phase transition was reported at around 8 T and 15 mK from the dHvA measurement, but the order parameter is unknown yet [28]. From the present $^{59}$Co-NMR measurement, it seems that the field-induced phase transition is not ascribed to magnetic in origin but would
Fig. 2. (a) The field dependence of $1/T_1T$ and ac magnetic susceptibility at 0.1 K. (b) The temperature dependence of $1/T_1T$ at several fields. The black arrow shows the $T_c$ at 4.22 T determined by the ac susceptibility measurement.

Fig. 3. (a) $^{59}$Co-NMR spectra measured at 0.1 K in 5.0, 5.2 and 5.4 T. (b) The temperature dependence of $^{59}$Co-NMR spectra at 5.2 T. Panels (a) and (b) show the Co-NMR center line arising from $|{-1/2}\rangle \leftrightarrow |{+1/2}\rangle$ transition.

-originate from the Fermi-surface instability probed by the transport measurement, since AFM magnetic fluctuations are suppressed at the higher fields [5]. However it is noted that the phase-transition was observed up to 9.5 T and seems to exist above 10 T in $H \parallel c$, which is anticipated to the relation with the magnetic Q phase observed in $H \perp c$. To understand the origin of the field-induced transition, further measurements, particularly angle dependence of the NMR measurements in the $ac$ plane down to 10 mK would give a clue to clarify the origin.
4. Conclusion

We have performed the single crystal $^{59}$Co-NMR measurements for $H \parallel c$ near $H_{c2}$ in CeCoIn$_5$ to investigate whether the field induced magnetic phase like the Q phase is observed or not. Although low temperature $1/T_1T$ shows a peak at 5.2 T above $H_{c2}$, $1/T_1T$ at 5.2 T is almost constant below 0.2 K, indicative of an absence of field-induced magnetic instability down to 0.1 K.

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