Neutron Diffraction in the Pressure-Induced Superconducting Antiferromagnet CeIrSi$_3$

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Abstract. Neutron diffraction experiments were performed to investigate a nature of the antiferromagnetic ordered phase below $T_N = 5.0$ K of the pressure-induced superconductor CeIrSi$_3$. We succeeded in observing incommensurate magnetic peaks characterized by the wave vector $\tau = (\pm 0.265, 0, 0.43)$. In contrast to the magnetic structure of CeRhSi$_3$, the observed magnetic structure is incommensurate both along the $a$ and $c$ directions. The antiferromagnetic ordered state in CeIrSi$_3$ could be interpreted as a spin-density wave formation.

1. Introduction
Coexistence between magnetism and superconductivity is the central issue in condensed matter physics. Pressure induced superconductivity discovered in CeRhSi$_3$ and CeIrSi$_3$ has attracted much attention due to their being heavy fermion superconductors without inversion symmetry. [1, 2, 3, 4, 5, 6, 7] It is also pointed out that both of them lie close to the quantum critical point, and interestingly Ce 4f electrons are itinerant even in their antiferromagnetic (AFM) phase in sharp contrast to CeRhIn$_5$ in which Ce 4f electrons are localized in the AFM phase. [8]

CeRhSi$_3$ and CeIrSi$_3$ crystallize in the same structure of the BaNiSn$_3$-type belonging to space group $I4mm$ (No. 107) without an inversion center. [9] Both materials show similar physical properties. [1, 3] They exhibit the AFM ordering below $T_N = 1.6$ K and 5.0 K at ambient pressure, respectively. By increasing the pressure, $T_N$ decreases and superconductivity appears in a wide pressure range from 1.2 to 2.7 GPa (and more) for both materials. The ground states of Ce ions in CeIrSi$_3$ and CeRhSi$_3$ are determined to be $\Gamma_7$ doublets through the susceptibility studies. [2, 3, 10]

The magnetic structure of CeRhSi$_3$ was determined by our previous study. [11] CeRhSi$_3$ shows an AFM phase below $T_N = 1.6$K with the magnetic moment of $\mu_{\text{ord}} = 0.13\mu_B$/Ce. The magnetic moments lie in the $c$ plane and form a longitudinal spin-density wave with a propagation vector $\tau = (0.215, 0, 1/2)$, being consistent with a picture that the Ce 4f electrons are itinerant. Concerning the magnetic structure for CeIrSi$_3$, there is only one neutron diffraction measurements on polycrystalline samples for CeIrSi$_3$, [12] which exhibits no magnetic reflections with possible maximum magnetic moment of 0.25 $\mu_B$/Ce. To determine the magnetic structure
which might be connected with the superconductivity in CeIrSi$_3$, therefore, we performed the neutron diffraction measurements using single crystalline CeIrSi$_3$.

2. Experimental

The single crystal sample used in the present study was grown by Czochralsky pulling method in a tetra-arc furnace at Tohoku University. The X-ray powder diffraction measurements at room temperature confirmed only the Bragg peak lines expected for the BaNiSn$_3$-type structure. We confirmed that the single crystal exhibits the AFM transitions in the wide $T$-$P$ phase diagram by the thermoelectric power and electrical resistivity measurements. [13] The size of the crystal was $\phi$ 3mm (at the maximum) in diameter and 35 mm long whose weight was 1.1 g.

Neutron diffraction measurements were carried out with the GPTAS triple-axis spectrometer installed at 4G in the JRR-3 research reactor at Japan Atomic Energy Agency. The incident momentum of $k_i = 3.83$ Å$^{-1}$ was adopted to avoid large absorption of Ir atoms. The spectrometer was operated by the configuration of 2-axis mode with a set of collimators of 40'-80'-40'. Pyrolytic graphite filters were utilized for both incident and scattered neutron to reduce the higher-order contaminations. The crystal was mounted to the $^3$He cryostat whose base temperature is 0.75 K. The lattice parameters at 1 K was $a^*=1.487$ Å$^{-1}$, and $c^*=0.642$ Å$^{-1}$.

3. Results and Discussion

To search the magnetic peaks in CeIrSi$_3$, we carried out survey scans along the major symmetry axes of two scattering planes of $(h0l)$ and $(hhl)$. Unfortunately we could not detect any magnetic Bragg signal in these scans. Therefore we focused on the $(h0l)$ reciprocal plane where magnetic Bragg reflections for CeRhSi$_3$ were observed. The $(h0l)$ reciprocal plane in CeTSi$_3$ ($T=$ Rh, Ir) is shown in Fig. 1 in which both nuclear and magnetic Bragg reflections are summarized for both materials. In the figure, the fundamental reflections for the BaNiSn$_3$-type structure are denoted by open circles, while magnetic Bragg reflections for CeRhSi$_3$ are denoted by black crosses.

![Figure 1. The $(h0l)$ reciprocal zone of CeTSi$_3$ ($T=$ Rh, Ir).](image)

To find the magnetic peaks in CeIrSi$_3$, we carried out a two-dimensional survey scans around the $Q = (0.215, 0, 2.5)$ at which the magnetic peak was observed in CeRhSi$_3$. As illustrated in Fig. 2, we discovered a weak intensity peak at an incommensurate reciprocal point of $Q = (0.265, 0, 2.43)$. Figure 2 (a) and (b) show typical scan profiles at $Q = (0.265, 0, 2.43)$ through (a) transverse and (b) longitudinal directions on the reciprocal plane. One can clearly recognize a single intensity peak in the plane at $T = 0.77$ K (below $T_N$), which disappears at
\( T = 8.0 \text{ K (above } T_N) \). These observations indicate that the Bragg reflections are of magnetic origin.

To confirm this scattering is real, we performed systematic survey scans at equivalent \( Q \) positions on the \((h0l)\) reciprocal plane. We could observe magnetic signals at 22 equivalent positions of \( Q = G \pm \tau_1 \) and \( Q = G \pm \tau_2 \) with \( \tau_1 = (0.265, 0, 0.43) \) and \( \tau_2 = (-0.265, 0, 0.43) \), and \( G \) denotes the fundamental reciprocal vector for the BaNiSn\(_3\)-type structure. In Fig. 1, observed magnetic Bragg satellites for CeIrSi\(_3\) in this study are also denoted by orange (for the propagation vector \( \tau_1 \)) and blue spots (for the propagation vector \( \tau_2 \)). In contrast to the magnetic structure of CeRhSi\(_3\), the observed magnetic structure is incommensurate both along the \( a \) and \( c \) directions.

Figure 3 show temperature dependence of the peak intensities at \( Q_1 = (0.265, 0, 2.43) \) (triangles) and \( Q_2 = (0.735, 0, 2.58) \) (crosses) in CeIrSi\(_3\), respectively. At \( Q_1 = (0.265, 0, 2.43) \), the intensities of 700 counts well below \( T_N \) gradually decreases with temperature and suddenly drops to the background intensity around \( T_N = 5.0 \text{ K} \). The temperature dependence at \( Q_2 = (0.735, 0, 2.58) \) also shows the similar behavior. It should be noted that the obtained Néel temperature of 5.0 K corresponds to the anomaly in both the resistivity measurements [3] and specific heat measurements [14] using single crystals.

The itinerant character of the Ce 4f electron in CeRhSi\(_3\) was pointed out by de Haas-van Alphen (dHvA) measurements. [2, 4, 5] The angle-resolved photoemission study in CeIrSi\(_3\) indicates that the Ce 4f electrons in CeIrSi\(_3\) are also itinerant. [15] The observed Fermi surfaces in CeIrSi\(_3\) are substantially larger than those in LaIrSi\(_3\) and they contact each other at Brillouin zone boundaries. Judging from the observed magnetic peaks and their propagation vector \( \tau = (0.265, 0, 0.43) \), the magnetic order is the spin-density wave polarized within the \( c \) plane, and the nesting of the Fermi surfaces would be its origin. These results strongly indicates that the Ce 4f electrons in CeIrSi\(_3\) as well as CeRhSi\(_3\) are itinerant, being consistent with other studies.

In conclusion, we succeeded in observing incommensurate magnetic peaks in CeIrSi\(_3\) below the ordering temperature \( T_N = 5.0 \text{ K} \). The magnetic structure of CeIrSi\(_3\) was characterized by the wave vector \( \tau = (0.265, 0, 0.43) \). It is very similar to that of CeRhSi\(_3\) \( \tau = (0.215, 0, 1/2) \), but it was incommensurate both along the \( a \) and \( c \) directions. The AFM ordered state in CeIrSi\(_3\) could be interpreted as a spin-density wave formation. We plan to perform quantitative analysis

![Figure 2](image-url)
Figure 3. Temperature dependence of magnetic peak intensity observed at $Q_1 = (0.265, 0, 2.43)$ (triangles) and $Q_2 = (0.735, 0, 2.58)$ (crosses) in CeIrSi$_3$. The intensities were subtracted by the background intensity at the paramagnetic phase.

with the data collected with the smaller size of the crystal with less absorption in the future.

Acknowledgements
The neutron experiments were supported by ISSP, University of Tokyo (PACS No. 9501 and 10501). N.A. was supported by a Grant-in-Aid for Scientific Research on Innovative Areas “Heavy Electrons” (No. 21102519, 23102722) from the Ministry of Education, Culture, Sports, Science and Technology, Japan (MEXT). N.A. is grateful to Dr. W. Higemoto (JAEA) for fruitful discussions.

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