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Neutron diffraction experiments have been performed on single crystalline samples of $CePtSi_3$. We found that the incommensurate magnetic propagation vector $\tau_1 = (\pm 0.283, 0, 0)$ in the phase I (2.2 ~ 4.8 K) switches to the commensurate vectors $\tau_2 = (\pm 0.25, 0, 0)$ and $\tau_3 = (\pm 0.25, 0, \pm 1)$ in the phase III (~ 2.8 K) at zero field. Although the magnetic structure of $CePtSi_3$ in the phase I is a spin density wave similar to those in $CeRhSi_3$, $CeIrSi_3$, and $CePdSi_3$, the magnetic structure in the phase III is commensurate unlike other $CeTSi_3$ (T = Rh, Ir, Pd) compounds. © *2018 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/).*
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I. INTRODUCTION

A finite antisymmetric spin-orbit interaction (ASOI) in the non-centrosymmetric systems lifts the electronic spin degeneracy without a magnetic field. The ASOI is one of the central issues in the condensed matter physics. In the strongly correlated *f*-electron physics, the effect of the ASOI on superconductivity and magnetism has attracted much attention since the discovery of the first heavy electron superconductor $CePt_3Si$ with a non-centrosymmetric crystal structure.[1] For example, pressure induced superconductors $CeRhSi_3$ and $CeIrSi_3$ show exotic superconducting properties such as the anomalously large upper-critical fields H_{c2}.[2,3] Neutron experiments for the non-centrosymmetric *f*-electron compounds were vigorously performed, but the definitive proof of the direct influence of the ASOI on magnetism is still absent.

Neutron diffraction experiments on $CeRhSi_3$ and $CeIrSi_3$ revealed that the magnetic moments of these compounds lie along the *a*-axis and form a longitudinal spin-density wave (SDW) with propagation vectors $\tau_{CeRhSi_3} = (0.215, 0, 0.5)$[4] and $\tau_{CeIrSi_3} = (0.265, 0, 0.43)$,[5] respectively. However, the magnetic fluctuations influenced by the ASOI have not been elucidated yet due to the large neutron absorption cross-section of Rh and Ir.

Here we focus on the non-centrosymmetric $BaNiSn_3$-type compounds $CePdSi_3$[6] and $CePtSi_3$.[7] The former exhibits successive magnetic transitions, weak ferromagnetism, and unusually complex metamagnetic transitions. The magnetic properties of the latter is very similar to those of $CePdSi_3$, but there are some qualitative differences. Unlike a weak ferromagnetic ground state observed in $CePdSi_3$, a proper antiferromagnetic (AFM) state may be realized in $CePtSi_3$. Although the origin of this remarkable difference is still unclear, we expect that the Rashba-type ASOI certainly influences their magnetic properties, and the strength of ASOI may cause the difference of magnetic behavior between $CePdSi_3$ and $CePtSi_3$.

As the first step of further understanding of the ASOI effect, we examined the magnetic structure of $CePtSi_3$ by single crystal neutron diffraction experiments.

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II. EXPERIMENTAL PROCEDURE

Single crystalline samples of $CePtSi_3$ were grown by a flux method just following the previous study.[7] The constituent elements (Ce, 3N; Pt, 3N; Si, 6N) of Ce:Pt:Si = 1:1:3 were premelted on a water-cooled copper hearth in an arc furnace to obtain a uniform solution. After that, the premelted product was put in an alumina crucible with the appropriate amount of tin as a flux. In order to examine the crystal structure, a number of single crystal samples were ground into powder and the powder X-ray diffraction pattern was measured at room temperature with Cu-K_α radiation using a commercial instrument (MiniFlex; Rigaku). The obtained diffraction pattern is consistent with the non-centrosymmetric body-centered tetragonal $BaNiSn_3$-type structure, and no impurity phase was observed within the experimental accuracy.

Neutron diffraction experiments were performed using the CORELLI elastic diffuse scattering spectrometer BL-9 and a cold neutron triple-axis spectrometer CTAX at Oak Ridge National Laboratory. In CORELLI, only a piece of single crystalline sample with size of $3 \times 3 \times 0.5$ mm^3 and weight of about 10 mg was attached to a Cu pin, and was cooled down with the base temperature of 0.25 K. On the other hand, several hundred of single crystalline samples (total amount is about 1 g) were glued on an Al plate by CYTOP (CTX-809X), and the measurements were carried out at temperature of 1.6, 2.5, 3.5, and 5.5 K on CTAX.

III. RESULTS AND DISCUSSION

Neutron diffraction experiments using the single crystalline sample of $CePtSi_3$ were performed to examine the nuclear and magnetic structures. Figure 1 shows the magnetic peak profiles around $(H, 0, 1)$ and $(H, 0, 2)$. In this figure, the black solid lines indicate the fitting curves with the Gaussian function. One can clearly recognize the magnetic Bragg reflections at incommensurate reciprocal point $Q_1 = (0.717, 0, 1)$ at 3.5 K and at commensurate points $Q_2 = (0.75, 0, 1)$ and $Q_3 = (0.75, 0, 2)$ below 2.5 K, respectively. Furthermore, the inset of Fig. 1(a) shows the magnetic peak profiles around $(H, 0, 0)$, where the 5.5 K data were subtracted as a paramagnetic contribution. In Fig. 1(a) and 1(b), shoulders can be seen on the right side of the magnetic peaks, but this is an artifact in the linewidth due to the several hundreds of single crystalline samples were coaligned. Because $Q_3 = (0.75, 0, 2)$ lies on the Brillouin zone boundary, it should be noted that two superlattice reflections are overlapped at Q_3. Taking this into account, we found that $CePtSi_3$ has propagation vectors $\tau_1 = (0.283, 0, 0)$, $\tau_2 = (0.25, 0, 0)$, and $\tau_3 = (0.25, 0, 1)$ at zero field, and the magnetic moment of $CePtSi_3$ does not lie on the a-axis but slightly tilted to the c-axis, unlike the longitudinal spin-density wave of $CeRhSi_3$[4] and $CeIrSi_3$.[5]

Figure 2 shows the temperature dependence of the magnetic peak intensities at $Q_1 = (0.717, 0, 1)$ (red), $Q_2 = (0.75, 0, 1)$ (blue), and $Q_3 = (0.75, 0, 2)$ (green), respectively. Since Q_3 lies on the Brillouin zone boundary, its intensity is a sum of two supperlattice reflections from the (101) and (103) zones, and the half of the intensity is plotted in Fig. 2. The *sum rule* of scattering intensity holds between $Q_1 = (0.717, 0, 1)$ and $Q_2 = (0.75, 0, 1)$ and $Q_3 = (0.75, 0, 2)$. At $Q_1 = (0.717, 0, 1)$, the intensity increases below $T_{N1} = 4.8$ K, decreases below 2.8 K and finally disappears at $T_{N2} = 2.2$ K. On the other hand, the intensities of $Q_2 = (0.75, 0, 1)$ and $Q_3 = (0.75, 0, 2)$ appear below 2.8 K and increase with lowering temperature. This behavior agrees with the anomaly in the preceding specific heat measurements, and neutron diffraction measurements revealed that the phase II corresponding to the temperature range from 2.2 K to 2.8 K was actually a transition region from the incommensurate to commensurate magnetic structures. The magnetic transition from incommensurate (SDW) order to commensurate order has also been reported for $CeCoGe_3$[8] and for $CeIrGe_3$.[9] However, the magnetic structure of the Ge-system is drastically different from those of the Si-system. The moment is parallel to the c-axis ($\mu // c$) in the Ge-system, whereas it lies within the ab-plane in the Si-system ($\mu \perp c$). Furthermore, the incommensurate structure consists of ferromagnetic sheets in the ab-plane which stack along the c-axis with various periodicities. It is unlikely that the nesting of the Fermi surface within the ab-plane causes incommensurate structures along the c-axis observed in the Ge-system. By contrast, the origin of SDW structure in $CePtSi_3$ is presumably due to the nesting of the Fermi surface.

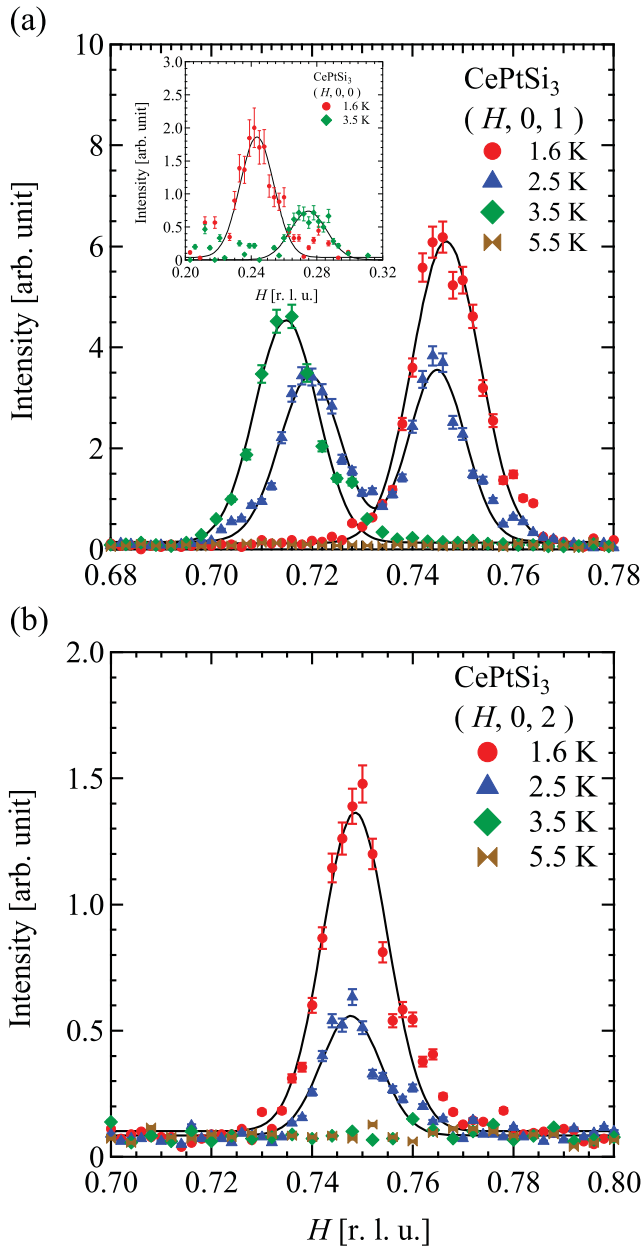

FIG. 1. Magnetic peak profiles around (a) (H, 0, 1) and (b) (H, 0, 2). The red circles, blue triangles, green diamonds, and brown symbols are the measurement data at 1.6, 2.5, 3.5, and 5.5 K, and black solid lines indicate the fitting curves with the Gaussian function. The inset of (a) shows the magnetic peak profiles around (H, 0, 0).

Finally, we consider the magnetic structure of $CePtSi_3$ with having these magnetic propagation vectors. In the phase I (2.2 ~ 4.8 K), the AFM ordered state in $CePtSi_3$ can be interpreted as a spin-density wave characterized by the wave vector τ_1 = (0.283, 0, 0), and it is schematically depicted in Fig. 3. Here, the red arrows show the direction and size of the cerium magnetic moments, and broken lines schematically represent a sinusoidal modulation of the magnetic moments. On the other hand, the magnetic structure in the phase III (~ 2.2 K) is commensurate with two propagation vectors τ_2 = (0.25, 0, 0) and τ_3 = (0.25, 0, 1). The size of ordered magnetic moment of $CePtSi_3$ is estimated to be 1.05(5) μ_B/Ce^{3+} at 4.0 K in this study. This value is slightly smaller than the value expected from the Γ_6 ground state (1.28 μ_B/Ce^{3+}), and this is partly because the observed temperature 4.0 K was relatively close to the transition temperature T_{N1} = 4.8 K and possibly the influence of the $c-f$ hybridization.

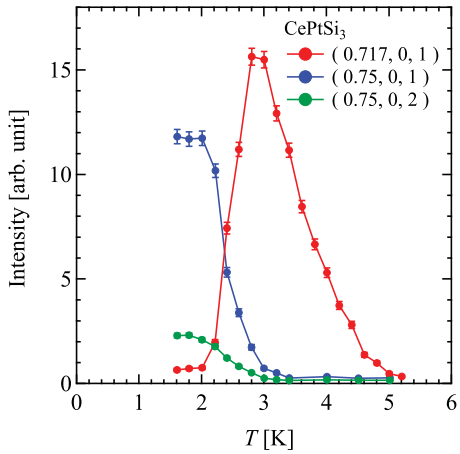

FIG. 2. Temperature dependence of the magnetic peak intensities observed at $Q_1 = (0.717, 0, 1)$ (red), $Q_2 = (0.75, 0, 1)$ (blue), and $Q_3 = (0.75, 0, 2)$ (green) in $CePtSi_3$.

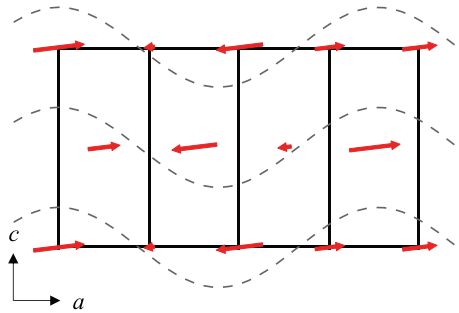

FIG. 3. Magnetic structure of $CePtSi_3$. The red arrows show the direction and size of the cerium magnetic moments.

In conclusion, we performed neutron diffraction experiments using a single crystalline sample of $CePtSi_3$ and examined the magnetic structure. We found that the incommensurate magnetic propagation vector $\tau_1 = (0.283, 0, 0)$ in the phase I ($\sim$ 4.8 K) switches to the commensurate vectors $\tau_2 = (0.25, 0, 0)$ and $\tau_3 = (0.25, 0, 1)$ in the phase III ($\sim$ 2.2 K) at zero field. Neutron diffraction experiments revealed that the phase II is actually a transition region from the incommensurate order in the phase I to the commensurate order in the phase III through the first order transition. Although the magnetic structure of $CePtSi_3$ in the phase I is a spin-density wave similar to those in $CeRhSi_3$, $CeIrSi_3$, and $CePdSi_3$, the magnetic structure in the phase III is commensurate, unlike other $CeTSi_3$ (T = Rh, Ir, Pd) compounds. So far there observed different magnetic structures in $CeTSi_3$ and $CeTGe_3$, but no clear evidence of the influence of ASOI has been elucidated. We plan to report quantitative analysis of neutron diffraction experiments under magnetic field in the near future.

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