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CROSS TOKAI **Experimental Report** J-PARC	承認日 Date of Approval 2017/07/20 承認者 Approver Takashi Ohhara 提出日 Date of Report 2017/07/19
課題番号 Project No. 2017A0223 実験課題名 Title of experiment Determination of the magnetic propagation wave vector of the non-centrosymmetric heavy electron compound CePdSi3 実験責任者名 Name of principal investigator Yoichi Ikeda 所属 Affiliation Institute for Materials Research, Tohoku University	装置責任者 Name of Instrument scientist Takashi Ohhara 装置名 Name of Instrument/（BL No.） BL18 (SENJU) 実施日 Date of Experiment 2017/05/12-2017/05/17 (5/12: setup&precooling) (5/13 13:30 ~ 5/17 9:00: on beam)

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。（適宜、図表添付のこと）

Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.

Sample information

Name: **CePdSi3 (single crystal)**

Dimension: ~ 2 mm x 2 mm x 0.05 mm-thickness

Bond: GE varnish, Holder: Cu sheet

Structural properties

Lattice constants: a = 4.331(2) Å, b = 4.331(2) Å, c = 9.61(4) Å, α = 90.0(2), β = 90.0(2), γ = 90.0(3), V = 180.3(7) Å^3 (which are evaluated from #8101-data measured at ~0.3 K).

Crystal structure: noncentrosymmetric body centered tetragonal

Space group: I4mm (#107)

Chemical formula: Ce Pd Si_3 (Z = 2)

Density: 6.094457 g cm^{-3}, Molecular weight: 330.792 g mol^{-1}

Physical properties

Magnetic transition temperatures: $T_{\rm I}$ = 4.8 K, $T_{\rm II}$ = 2.8 K, $T_{\rm III}$ = 2.3 K

Neutron scattering lengths and cross sections (cited from the NIST data base):

Atom	b / fm	σ_{abs} / barn	σ_{inc} / barn	At.wt. / g mol^{-1}
Ce	4.840	0.63	0.0	140.116
Pd	5.910	6.90	0.093	106.420
Si	4.149	0.171	0.004	28.0855

Ref. [1] D. Ueta et al., JPSJ 85, 104703 (2016).

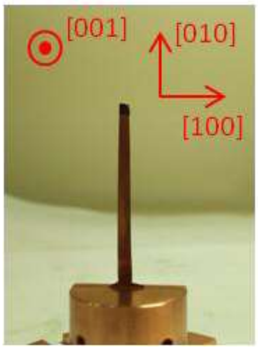

Fig. 1 Photograph of the single crystalline sample of $CePdSi_3$ and the Cu holder for a ^{3}He refrigerator.

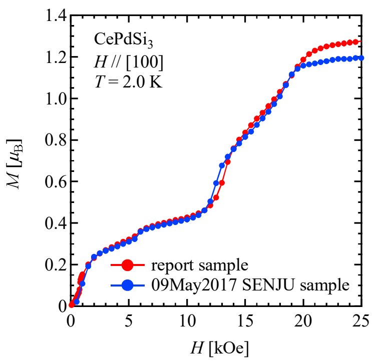

Fig. 2 Magnetization curves of $CePdSi_3$. The sample quality of this experiment is in good agreement with that of the reported sample [1].

2. 実験方法及び結果（実験がうまくいかなかった場合、その理由を記述してください。）

Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.

[Experimental procedure]

$CePdSi_3$ shows three successive magnetic transitions at T_{I} = 4.8 K, T_{II} = 2.8 K, and T_{III} = 2.3 K in zero magnetic field. In order to determine the magnetic propagation vector in the three magnetic phases, we carried out a neutron diffraction experiment with the SENJU diffractometer at BL18. To investigate the magnetic structure below T_{III} (phase III), we utilized a ^{3}He refrigerator and cooled the sample down to ~ 0.3 K. The equator scattering plane was set into the (*h*0*l*) plane in reference to the experimental procedure for similar noncetrosymmetric $CeTSi_3$ systems. In this experiment, we used a radial collimator system on BL18 to reduce background and spurious signals from the cryostat. The accelerator was operated at 150 kW, and there is no serious (long time) beam-supply stop in our experiment. From the results of premeasurements (#8101-8104), data acquisition times were appropriately determined so that we obtain a data set with a sufficient statistical quality. The experimental history is summarized in Fig. 3.

2017	DAQ				gonio	chopper	temperature controller			remarks
date	run#	kicker#	time [m]	time [h]	w	frame	T_sorb	T_samp	phase	
13-May	8098	–	–	–	-10	1st	2.7 oneshot	BT(0.3)	III	
	8099	–	–	–	–	–	2.7 oneshot	BT(0.3)	III	alignment
	8100	–	–	–	–	–	2.7 oneshot	BT(0.3)	III	
	8101	306000	204.0	3.4	89	1st	2.7 oneshot	BT(0.3)	III	search for mag. peaks at BT
	8102	153000	102.0	1.7	89	2nd	2.7 oneshot	BT(0.3)	III	
	8103	153000	102.0	1.7	71.5	2nd	2.7 oneshot	BT(0.3)	III	search for mag. peaks at BT, covering the dead ang.
	8104	306000	204.0	3.4	71.5	1st	2.7 oneshot	BT(0.3)	III	
14-May	8105	1224000	816.0	13.6	89	1st	2.7 oneshot	BT(0.3)	III	overnight scan at BT for improving statistics
	8106	776826	517.9	8.6	89	2nd	2.7 oneshot	BT(0.3)	III	
15-May	8107	301702	201.1	3.4	89	1st	23.5~23.25	1.45(5)	III	check for temp. variation of tau below TIII
	8108	159029	106.0	1.8	89	2nd	23.5~23.25	1.45(5)	III	
	8109	48321	32.2	0.5	89	1st	41 condense	2.55(5)	II	
	8110	–	–	–	–	–	–	–	–	
	8111	768000	512.0	8.5	89	1st	41 condense	2.55(5)	II	search for mag. peaks in phase II
	8112	315926	210.6	3.5	89	2nd	41 condense	2.55(5)	II	
	8113	142371	94.9	1.6	89	2nd	41 condense	2.55(5)	II	
16-May	8114	306000	204.0	3.4	89	1st	50 condense	5.0(1)	PM	background measurement in the PM phase
	8115	157625	105.1	1.8	89	2nd	50 condense	5.0(1)	PM	
	8116	459000	306.0	5.1	89	1st	50 condense	3.5(1)	I	
	8117	459000	306.0	5.1	89	2nd	50 condense	3.5(1)	I	search for mag. peaks in phase I
	8118	517501	345.0	5.8	89	1st	50 condense	3.5(1)	I	

Fig. 3 History of our experiment at BL18. The label of I, II, and III means the phase I (T_{I} ~ T_{II}), II (T_{II} ~ T_{III}), and III (< T_{III}), respectively.

[Experimental Results]

In the paramagnetic phase ($T > T_{\mathrm{I}}$), the crystal structure of $CePdSi_3$ was confirmed to be a noncentrosymmetric body-centered tetragonal structure (space group I4mm; #107), for which nuclear Bragg reflections have an extinction rule of $h + k + l = 2n$ (*n*: integer). For example, nuclear Bragg reflections in the (1*kl*) plane are observed at $k + l$ = odd, as shown in Fig. 4(a). Here the observed nuclear Bragg reflections are indicated with blue circles. Note that (1-10), (11-2), and (1-1-2) reflections in Fig. 4(a) are out of the observable range for this experiment. The surveyable area in the (1*kl*) plane is indicated with the yellow-hatched area in Fig. 5. Lattice constants at 0.3 K were determined as a = 4.331(2) Å, b = 4.331(2) Å, c = 9.61(4) Å, α = 90.0(2), β = 90.0(2), γ = 90.0(3), V = 180.3(7) Å^3, while other parameters (position parameter *z*) are under analysis.

2. 実験方法及び結果（つづき）Experimental method and results (continued)

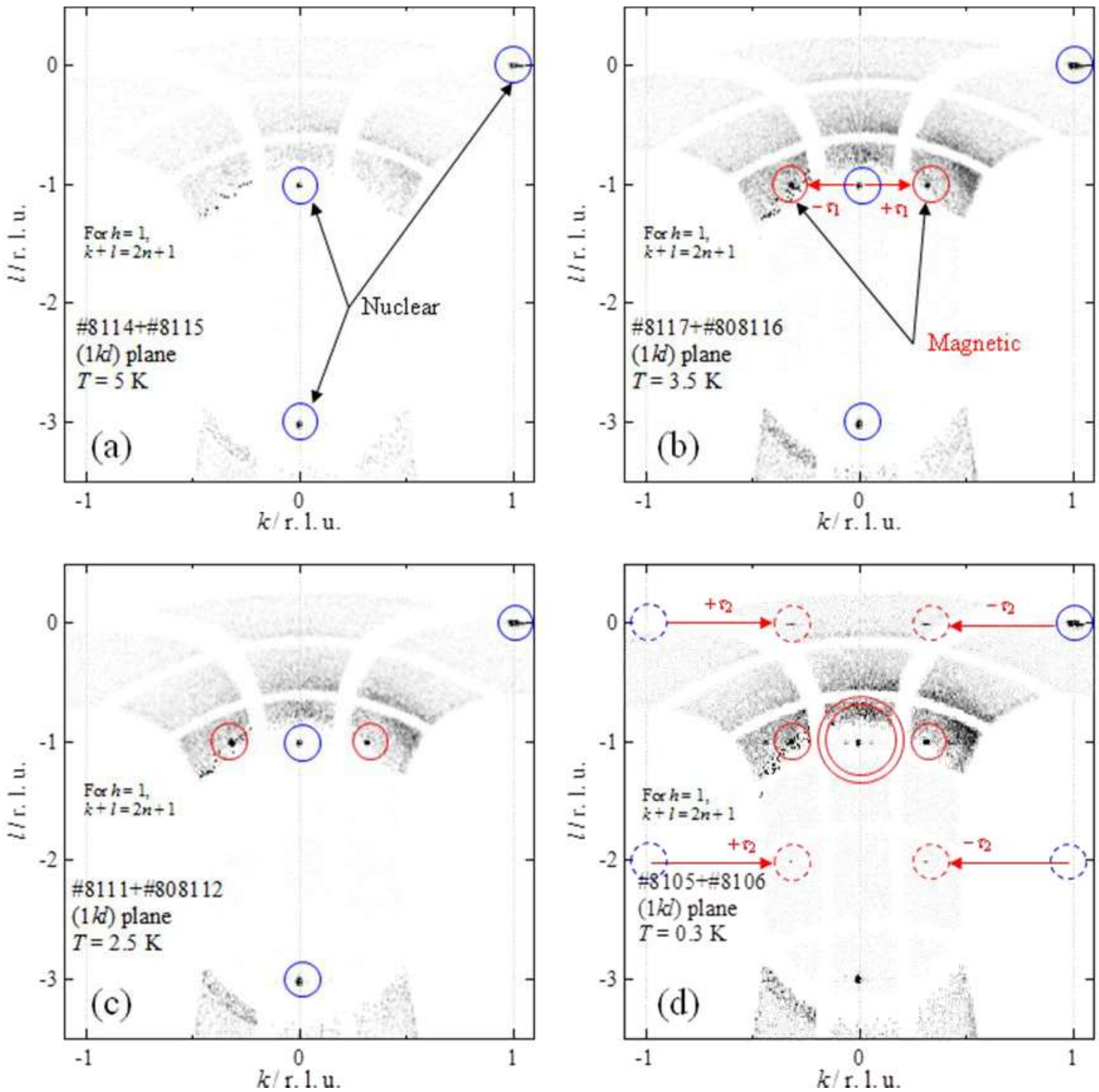

Fig. 4 Neutron intensity maps of $CePdSi_3$ on the (1*kl*) plane measured at (a) 5 K, (b) 3.5 K, (c) 2.5 K, and (d) 0.3 K.

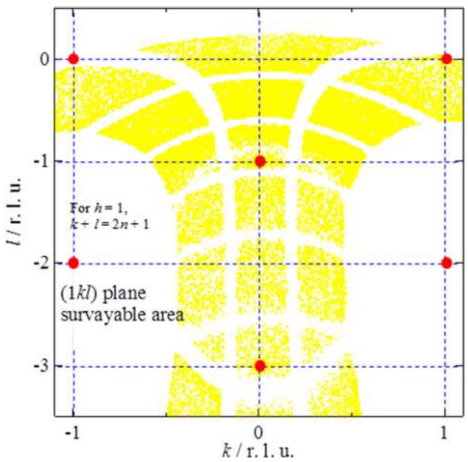

Fig. 5 Surveyable reciprocal space in the (1*kl*) plane. The yellow-hatched area indicates the surveyable area for this experiment. The filled-red circles show fundamental nuclear Bragg reflections in the (1*kl*) plane for I4mm space group. In this experiment, (1-10), (11-2), and (1-1-2) reflections are out of range.

2. 実験方法及び結果（つづき）Experimental method and results (continued)

In the magnetic phase between T_{I} = 4.8 K and T_{II} = 2.8 K (phase I), magnetic Bragg reflections were observed at several incommensurate positions described with the magnetic propagation wave vector of τ_1 = (0.314, 0, 0) as shown in Fig. 4(b). Here the magnetic Bragg reflections are indicated with red circles, and the propagation vector τ_1 is indicated with red arrows. Note that the magnetic reflections at around ***Q*** = (1, 1, 0) and (1, 0, -3) in Fig. 4(b) are also out of range although those reflections are equivalent with that around ***Q*** = (1, 0, -1). In the magnetic phase between T_{II} = 2.8 K and T_{III} = 2.3 K (phase II), neutron intensity map is almost the same as that of the phase I, as seen in Fig. 4(c). In the magnetic phase below T_{III} = 2.3 K (phase III), additional magnetic reflections were observed at several incommensurate positions described with τ_2 = (0.686, 0, 0) as shown in Fig. 4(d), here the magnetic reflections of τ_2 are indicated with broken-red circles. Furthermore, significant weak satellite reflections were observed at around nuclear Bragg positions. For example, these satellites around ***Q*** = (1, 0, -1) are indicated with double circles in Fig. 4(d). An enlarged view around ***Q*** = (1, 0, -1) is shown in Fig. 6(left). The observed satellite peaks seems to be a 3*Q*-harmonics for τ_2. Additional fragile spots around (1, ±0.45, -1) are under analysis.

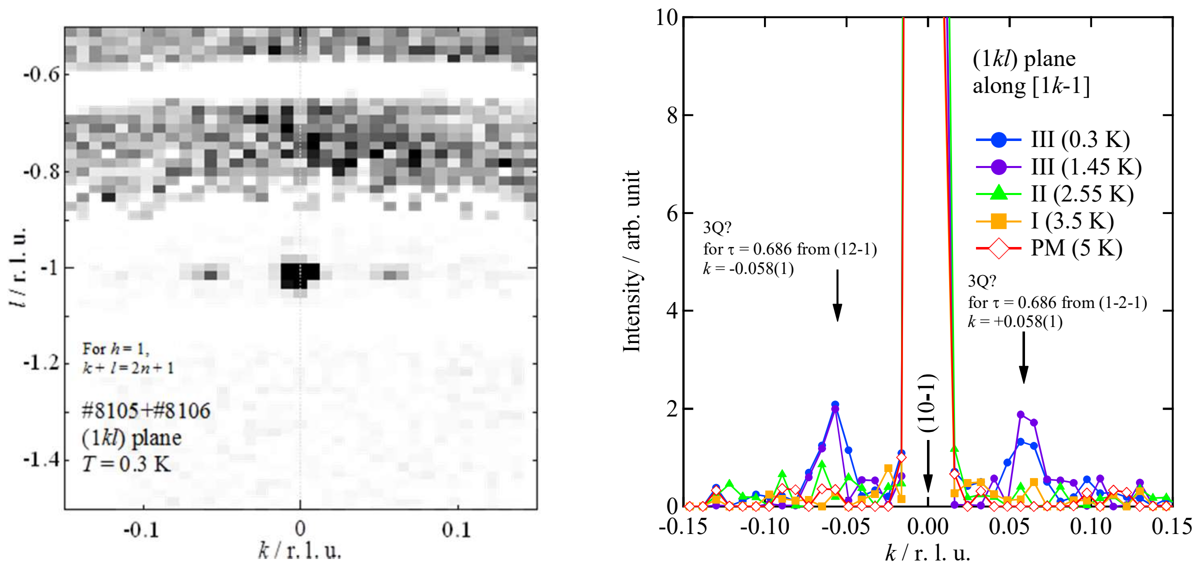

Fig. 6 (left) An enlarged view around ***Q*** = (1, 0, -1) in Fig. 4(d). (right) Line profiles around ***Q*** = (1, 0, -1) along the *k*-direction measured.

IR	BV	Atom	BV components					
			$m_{\parallel a}$	$m_{\parallel b}$	$m_{\parallel c}$	$im_{\parallel a}$	$im_{\parallel b}$	$im_{\parallel c}$
Γ_1	ψ_1	1	0	2	0	0	0	0
Γ_2	ψ_2	1	2	0	0	0	0	0
	ψ_3	1	0	0	2	0	0	0

Table 1. A result of the representation analysis with "SARAh" software. Basis vectors for I4mm with τ_1 = (0.314, 0, 0) or τ_2 = (0.686, 0, 0) are listed. The decomposition of the magnetic representation for the Ce site (0, 0, 0) is $\Gamma_{mag} = 1\Gamma_1^1 + 2\Gamma_2^1$.

2. 実験方法及び結果（つづき） Experimental method and results (continued)

From a result of the representation analysis with "SARAh" software, there is two possible magnetic structures (Γ_1 and Γ_2) for τ_1 = (0.314, 0, 0) and τ_2 = (0.686, 0, 0) as shown in Table 1. For Γ_1, magnetic moments are perpendicular to the propagation vector τ_1, indicating a transverse sinusoidal magnetic structure (*m* // *b*-axis ⊥*a*-axis). On the other hand, Γ_2 is described by a linear combination of the two basis Ψ_2 and Ψ_3, $c_2\Psi_2 + c_3\Psi_3$, so that Γ_2 allows the introduction of an out-of-plane (*c*-axis) component. For example, for $c_3 = 0$, magnetic moments have no *c*-axis component suggesting a longitudinal (sinusoidal) magnetic structure (*m* // *a*-axis). While, for non-zero c_3, a longitudinal (sinusoidal) magnetic structure with a finite out-of-plane component can be realized. The case of $c_2 = 0$ (transverse magnetic structure; *m* // *c*-axis) may be excluded from the results of the previous magnetization measurements [1], because the *c*-axis of $CePdSi_3$ is the magnetic hard axis.

A weak ferromagnetic component was observed below T_{II} from the magnetization measurement [1], so that the Γ_1 magnetic structure may drop out as a candidate of the magnetic structure of the phase II, and III. In this case, it is expected that neutron diffraction intensity at fundamental nuclear Bragg positions depends on temperature. Temperature dependence of the neutron intensity is under analysis.

In conclusion, we carried out a single-crystal neutron diffraction experiment to investigate the magnetic structure of a noncentrosymmetric compound $CePdSi_3$ by utilizing the SENJU diffractometer at BL18, J-PARC. Clear incommensurate magnetic Bragg reflections were observed at τ_1 = (0.314, 0, 0) in the phase I and II. In addition, additional Bragg peaks were observed at τ_2 = (0.686, 0, 0) below T_{III} (phase III). From the results of the magnetization measurements and the representation analysis, we proposed an incommensurate longitudinal sinusoidal magnetic structure (Γ_2: *m* // *ac*) as a possible magnetic structure in the phase I (T_I = 4.8 K < T < T_{II} = 2.8 K). The *c*-axis ferromagnetic component is considered to be zero in the phase I from the results of the magnetization measurements. On the other hand, a weak ferromagnetic component was observed below T_{II}. Therefore, an incommensurate longitudinal sinusoidal magnetic structure with a finite (uniform) *c*-axis component may be realized in the phase II. In the phase III, magnetic propagation wave vector is changed to τ_2 from τ_1. Since two inequivalent magnetic Bragg reflections were observed in the phase III, a phase separating state between τ_1 and τ_2 phases (or a weak structural transition) may occur at least at 0.3 K. Further precise analysis is indispensable for publishing.