CROSS TOKAI Experimental Report J-PARC	承認日 Date of Approval 2016/1/18 承認者 Approver Takashi Ohhara 提出日 Date of Report 2016/1/15
実験課題番号 Project No. 2014P0203 実験課題名 Title of experiment Structure and electronic properties of functional materials 実験責任者名 Name of principal investigator Ryoji Kiyanagi 所属 Affiliation J-PARC neutron science section	装置責任者 Name of responsible person Takashi Ohhara 装置名 Name of Instrument/(BL No.) SENJU/BL18 利用期間 Dates of experiments 2014/6/6-2014/6/14 2015/3/29-2015/4/1

1．研究成果概要（試料の名称、組成、物理的・化学的性状を明記するとともに、実験方法、利用の結果得られた主なデータ、考察、結論、図表等を記述してください。

Outline of experimental results (experimental method and results should be reported including sample information such as composition, physical and/or chemical characteristics.

1. Crystal and magnetic structure of $SmBaMn_2O_6$

The materials represented as $ReBaMn_2O_6$ (Re = trivalent rare-earth metal) exhibit variety of orderings due to the competition between charges, orbitals and spins, as is seen in $ReMnO_3$ type materials. $ReBaMn_2O_6$ has a double perovskite-type structure with the A-sites alternately occupied by Re and Ba ions along *c*-axis. In the case of Re = Sm, a charge-orbital ordered phase (COO1) emerges below 380 K followed by a re-arrangement of COO (COO2) around 200 K accompanied by an antiferromagnetic phase transition. In addition, an anomaly in the magnetization around 10 K has been reported. Although the structure and the charge-orbital (CO) ordering pattern have already been understood, the magnetic structure and the relation between the re-arrangement of the CO ordering and the magnetic ordering around 200 K have not been clarified, yet. In this experiment, neutron single crystal diffraction was conducted on SENJU in order to study the magnetic structure and the details of the phase transitions.

The experiment was conducted on the single crystal neutron diffractometer, SENJU, at BL18 in MLF. A single crystal of $SmBaMn_2O_6$ with the size of ~ 2.0 x 4.0 x 1.2 mm^3 was mounted on a cryostat. The measurements were performed around 200 K, 10 K and varying the temperature to study the evolution of superlattice reflections.

The observed diffraction pattern clearly revealed the existence of two types of magnetic superlattice reflections, one is at (0 1/2 1/2) and the other (1/2 1/2 1/2), as seen in Fig. 1. These propagation vectors coincide with the CO ordering patterns of Mn ions. The propagation vectors were found to be unchanged below 10 K, while the changes in intensities of the magnetic superlattice reflections were observed. This suggests that only the orientations of the spins are changed below 10 K. As for the phase transitions around 200 K, the temperature dependences of the superlattice reflections of COO1 and the magnetic reflections clarified that the transformation

of COO1 to COO2 occurred just above the antiferromagnetic phase transition, being indicative that the re-arrangement of CO ordering evokes the antiferromagnetic phase transition (Fig. 2).

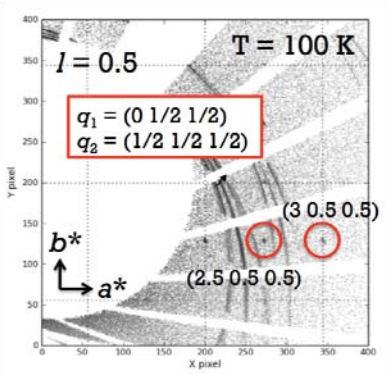

Fig. 1 Observed magnetic superlattice reflections at q_1 = (0 1/2 1/2) and q_2 = (1/2 1/2 1/2)

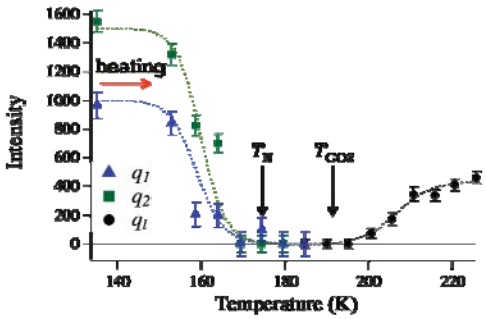

Fig. 2 Temperature dependence of the observed suplerlattice reflections

2. Magnetic structure analysis of Pt–Mn alloy

Sample; $Pt_{1-x}Mn_x$ (x = 0.14) , 12mmϕ, 6mmh, a rod-shape single crystal

Introduction; Atomic and magnetic ordered structures of $Pt_{1-x}Mn_x$ (x = 0.14) were studied as a series of Pt-rich Pt-Mn alloys with ABC_6-type ordered structure. When the complete ABC_6-type order is formed in the alloys, a long-ranged anti-ferromagnetic order of type III in the fcc lattice is formed. The ABC_6-type ordered structure has the largest unit cell in the fcc based ordered structures and is only found in Pt–rich Pt–Mn, Pt-rich Pt-Cu and ternary $CuMnPt_6$ alloys, and Pt-Mn alloys are the only example that forms magnetic order on the ordered structure. In the present study, detailed structure of both magnetic and atomic orders have been investigated quantitatively.

Experimental method; In the experiment at BL18, nuclear Bragg intensities were collected by the 1st. frame measurements to obtain order parameters for the ABC_6-type ordered structure, and magnetic scattering in the (hk0) reciprocal plane was observed by the 2nd. frame measurements.

Results; Figure 1(a) shows plots of nuclear intensities at fundamental and superlattice points (X and L points of fcc fundamental lattice) as a function of the reciprocal vector q (Å). By analyzing their intensity ratios, the order parameters for the ABC_6-type ordered structure are deduced to be $S_1 = S_2 = 1.0$, showing complete atomic order is formed in the alloy. Figure 1(b) shows intensity distribution of the magnetic scattering in the (hk0) plane at 6.7K. Anti-ferromagnetic superlattice reflections of type-III order are observed at -1 -3/2 0 and its equivalents. Their integrated intensities are plotted against temperature in Fig. 1(c). The temperature variation show that antiferromagnetic transition takes place at T_N = 20 K in the alloy. Using structural parameters obtained in the analysis of atomic structure, the integrated intensities of magnetic reflections are now being analyzed with a model of longitudinal spin alignment on the type-III anti-ferromagneticorder as shown in Fig.2. The present

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study will clarify the precise magnetic structure formed on the ABC_6-type ordered structure.

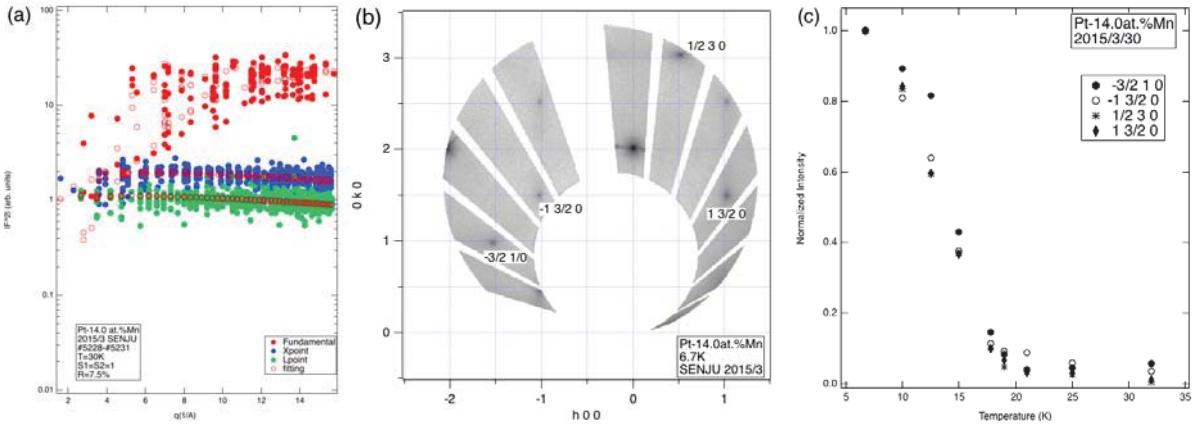

Fig.1 (a) Intensity versus q (Å) plots for fundamental (red closed circles) and superlattice points, X and L points of fcc lattice (closed circles of blue and green, respectively) measured by 1.st frame setting, with result of fitting (red open circles) (b) Intensity distribution in the (hk0) plane at 6.7K, with magnetic reflections. (c) Temperature variation of magnetic reflections at -3/2 1 0 (●), -1 -3/2 0 (○), 1/2 3 0 (※), and 1 3/2 0 (♦).

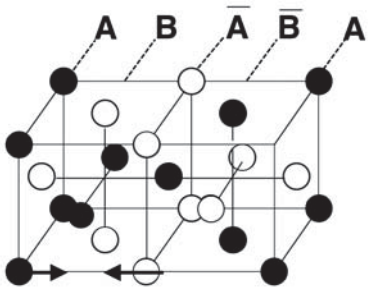

Fig.2：Model of type-III magnetic structure with longitudinal spin alignment.