MLF Experimental Report	提出日 Date of Report 2013/10/3
課題番号 Project No. 2012B0226 実験課題名 Title of experiment High Magnetic Field Neutron Diffractions in Frustrated Multi-ferroics 実験責任者名 Name of principal investigator Hiroyuki Nojiri 所属 Affiliation Institute for Materials Research, Tohoku University	装置責任者 Name of responsible person Kenichi Oikawa 装置名 Name of Instrument/（BL No.） BL10 実施日 Date of Experiment 2013/04/03-2013/04/12

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.
$MnWO_4$, Single Crystal

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.
The target of this proposal is the multi-ferroic sample $MnWO_4$, which shows the memory effect in high magnetic field. It consists of zig-zag chains running along the *c*-axis. For the strong spin frustration, three different magnetic phases;AF1(uudd), AF2 (non-collinear, incommensurate cycloidal), AF3 (incommensurate sinusoidal) appear at zero field. There are at least three phases in high magnetic fields; HF, IV and V as shown in Fig. 1. When a magnetic field is applied along the easy-axis, a distinct memory effect is found. The key experiment to understand the mechanism of the multi-ferroic behavior and the distinct memory effect in $MnWO_4$ is the determination of magnetic structures in the high magnetic field phases HF, IV and V. In the previous neutron diffraction up to 30 T along the scan A and B in the Figs. 1 and 2 made at SNS, Oakridge, we found that the propagation vector is slightly different between the phase IV and the AF2. The splitting of commensurate peak is also found in HF phase. The next step is to investigate the reentrant transitions between HF and the phase IV and between the phase IV and the phase V. These phase boundaries are determined by the macroscopic measurement and thus it is not clear if the phase IV persists to the low temperature or not. If the phase V is the paramagnetic or not is also unclear yet. The neutron diffraction in 40-50 T range would give us the clear answer to this points.

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

The proposal would have a strong impact to understand the full magnetic phase diagram of $MnWO_4$ because it is the most established model system of multi-ferroics. In the present experiment, we have scanned along the orange arrow in Fig. 1

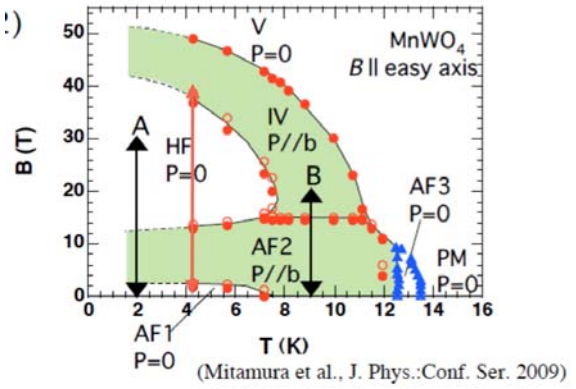

Fig. 1 Proposed phase diagram of $MnWO_4$. The orange arrow shows the scan in this experiment.

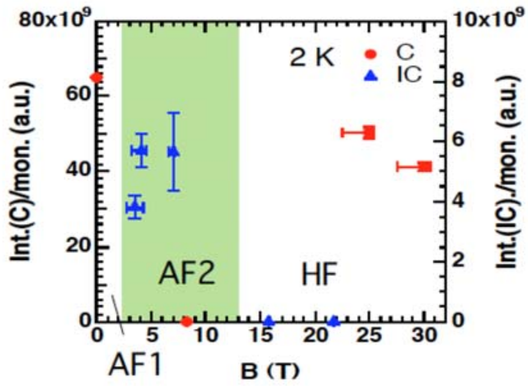

Fig. 2 Magnetic field dependence of intensities of two components measured at SNS. C and IC stand for commensurate and incommensurate peaks, respectively.

Figure 3 shows the example of TOF and magnetic field curve. In this setting, the TOF of the incommensurate peak is set at the magnetic field peak by the adjustment of the delay between the T_0 of neutron beam and the pulsed magnetic field. At the TOF of C-peak, the magnetic field is as low as 33.3 T. In a single setting of delay, we can trace the two peaks. For example, C-peak disappears when the delay is adjusted and the 40 T peak is located at the TOF of C-peak. By adjusting the peak field of the pulsed magnet and the time delay, we have measured the magnetic field dependence of two components up to 40 T.

Figure 4 shows the phase diagram made from the present results. The alternation of the commensurate and the incommensurate structures is found at around 38 T. It is the direct evidence of the second reentrant phase boundary in this system. It is also noteworthy that there is still substantial intensity in the incommensurate peak. This shows that the phase above the second phase boundary is not a forced ferromagnetic phase.

To explain the complicated phase diagram of this compound, we are working with model calculations. To have such second reentrant phase boundary, we need either compeating anisotropies or bi-quadratic interactions.

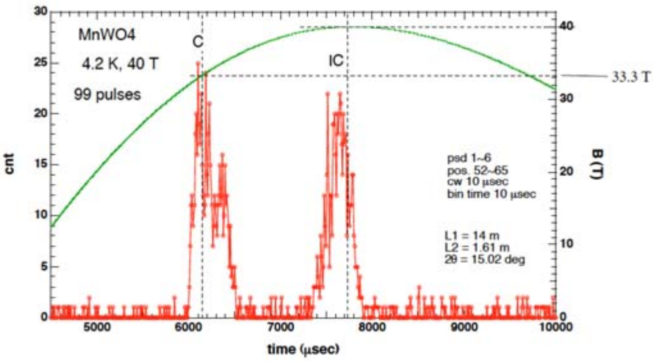

Fig. 3 The TOF and field trace

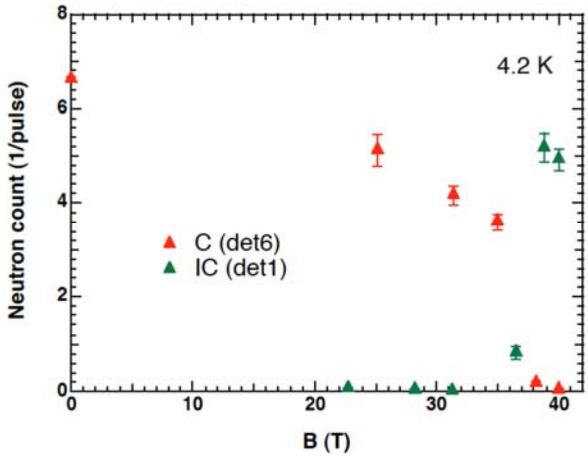

Fig. 4 Magnetic field dependence of intensities of two components taken at J-PARC

In summary, we have succeeded in determining the second reentrant phase boundary of $MnWO_4$ by using the neutron diffraction in pulsed magnetic fields up to 40 T. The major part of the phase diagram of this compound is determined by direct diffraction experiments. The present technique would be useful to investigate varieties of compounds with complicated magnetic phase diagrams.