


(※本報告書は英語で記述してください。ただし、産業利用課題として採択されている方は日本語で記述していただいても結構です。)

	承認日 Date of Approval 2017/07/14 承認者 Approver Ryoichi Kajimoto 提出日 Date of Report 2017/07/13
課題番号 Project No. 2014B0152 実験課題名 Title of experiment Identifying spin and orbital excitations in a spinel vanadate 実験責任者名 Name of principal investigator Hajime Sagayama 所属 Affiliation Photon Factory, IMSS, KEK	装置責任者 Name of responsible person Ryoichi Kajimoto 装置名 Name of Instrument/(BL No.) 4SEASONS/BL01 実施日 Date of Experiment 2015/4/9 11:00 - 4/13 11:00

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.

Chemical formula of the sample is  $MnV_2O_4$ . Single crystals were grown using the floating-zone method. They were characterized by powder x-ray diffraction and magnetization measurements. Four crystal bars, having a total mass of 10g, were coaligned using backscattering Laue photographs. The sample was also used in 2012A

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 method*

Two-dimensional detectors and multiple  $E_i$ 's enable the efficient measurement of large volumes of reciprocal space at BL01. The incident energies of the neutrons were selected as 73, 34, 19, and 12 meV by a Fermi chopper. The sample was rotated around [001] by a 0.5-deg. step per 15 min. The temperature was maintained at 5.6 K. Using the software UTSUSEMI, we extracted the value proportional to the dynamical structure factor. In addition, the intensity was divided by the Bose factor. The binning parameters were 0.1 (r.l.u.) for H, K, and L and 0.4 meV for the energy.



Figure 1. Single crystals of  $MnV_2O_4$  used in this experiment

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

### Experimental results

Figures 2 show the representative experimental results of the INS at  $E_i=34$  meV and  $T=5.6$  K along three high symmetry axes: (a)[hhh], (b)[hh0], and (c) [h00]. The corresponding calculations with Heisenberg-type spin Hamiltonian are shown in Figs. 2(d)–2(f). Calculated intensity are averaged those from a multiple-domain sample for comparison with the experimental results. The Calculation reproduces the experimental result in energy region below 17 meV. In Fig. 2(a), we observed not only an acoustic branch from the  $\Gamma$  point (111) but also a clear optical branch with an 8–9 meV energy gap. In our calculation, the former and the latter branches correspond to the  $\text{Mn}^{2+}$  acoustic and optical modes, respectively. In addition, the dispersive excitations around 10 meV is newly observed along [hh0] [Fig. 2(b)]. Figure 2(g) shows the energy-intensity plot at [hh0] ( $h=1.8$ – $2.2$ ), at  $E_i=19$ meV. An inelastic scattering peak with a width of  $\sim 3$ meV is observed around 10 meV. Considering the energy resolution to be less than 1 meV, this broad peak may be ascribed to at least two branches. Our calculation suggests that the lower- and higher-energy branches should be assigned to the  $\text{Mn}^{2+}$  optical mode and  $\text{V}^{3+}$  mode, respectively. In Figs. 2(b) and 2(c), we can see a dispersive mode with a large scattering intensity around 20 meV at both (2,2,0) and (4,0,0). This scattering cannot be explain by our calculation. Figures 2(h)–2(q) illustrate the constant energy cut of the scattering intensity around (2,2,0) and (4,0,0). The scattering around (4,0,0) [Figs. 2(m)–2(q)] seems to be a typical Heisenberg-type spin wave excitation as well as  $\text{Mn}^{2+}$  acoustic mode [Figs. 2(h)–2(l)]. This result shows that we have to consider another theoretical model including the spin-orbit interaction.

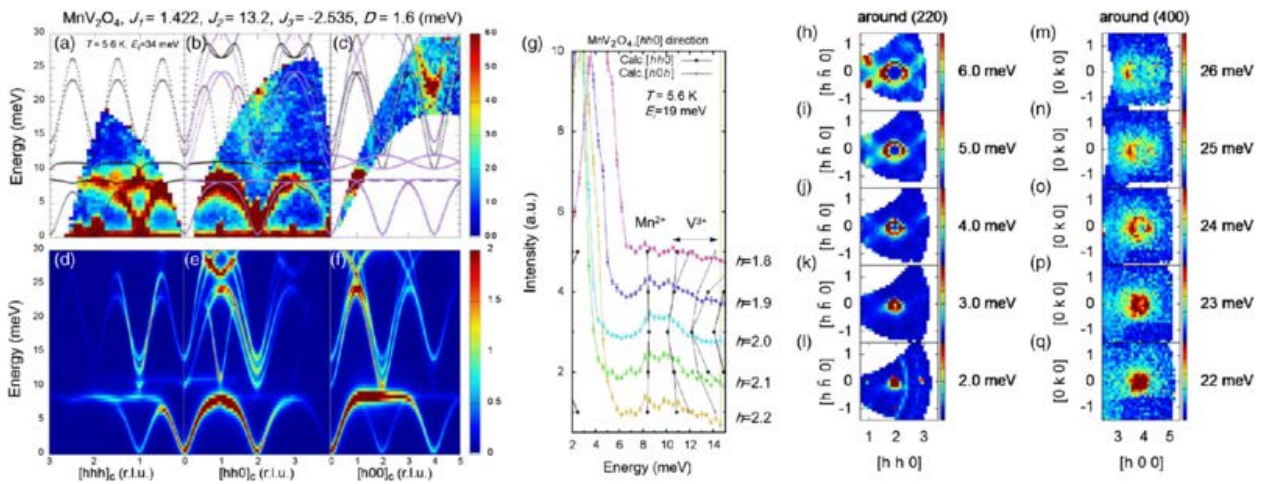


Figure 2. Experimental inelastic intensity (a)–(c) and the corresponding calculated intensity (d)–(f) contour along the three high symmetry axes. (g) Plot of inelastic scattering intensity around [220]. (h)–(k) Inelastic intensity contour in the [hh0]–[h $\bar{h}$ 0] plane around [220]. (m)–(q) Inelastic intensity contour in the [h00]–[0k0] plane around [400].