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	承認日 Date of Approval 2017/7/14 承認者 Approver Ryoichi Kajimoto 提出日 Date of Report 2017/7/13
課題番号 Project No. 2012A0058 実験課題名 Title of experiment Spin and orbital excitations in spinel vanadate 実験責任者名 Name of principal investigator Hajime Sagayama 所属 Affiliation Dept. of Adv. Mat. Sci., Univ. of Tokyo	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 2012/11/8 11:00~11/12 10:00

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.

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.
<p><i>Experimental method</i></p> <p>Two-dimensional detectors and multiple <math>E_i</math>'s enable the efficient measurement of large volumes of reciprocal space at BL01. The incident energies of the neutrons were selected as 153, 63, and 34, meV by a Fermi chopper. The sample was rotated around [001] by a 1.5-deg. step per 60 min from <math>\phi = -42</math> to 30 deg.. (When <math>\phi = 0</math>, the [100] direction was parallel to the incident neutron beam.) In addition, it was also rotated by a 3-deg. step per 60 min from 30 to 66 deg.. 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.</p>



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 inelastic neutron scattering at  $E_i=34$  meV and  $T=5.6$ K along three high symmetry axes,  $[hhh]$ ,  $[hh0]$ , and  $[h00]$ . 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. The corresponding calculation result with Heisenberg-type spin Hamiltonian are demonstrated as green and pink lines in the fig. 2. It reproduces well the experimental result in energy region below 10 meV. In our calculation, the former and the latter branches correspond to the  $Mn^{2+}$  acoustic and optical modes, respectively. Our calculation suggests existence of one  $V^{3+}$  spin acoustic mode and three optical mode above 10 meV. However, unfortunately, we could not find obvious magnetic excitations above 10 meV. Since cubic structural symmetry is lowered to tetragonal by the orbital ordering in  $MnV_2O_4$ , the neutron scattering intensities from the three directional domains are averaged. The intensities may spread, probably because dispersion relations of the  $V^{3+}$  spin excitations are anisotropic. In order to confirm those, we have to conduct inelastic neutron scattering experiment with single domain sample. It can be realized by applying a magnetic field or uniaxial pressure along the  $[100]$  direction.

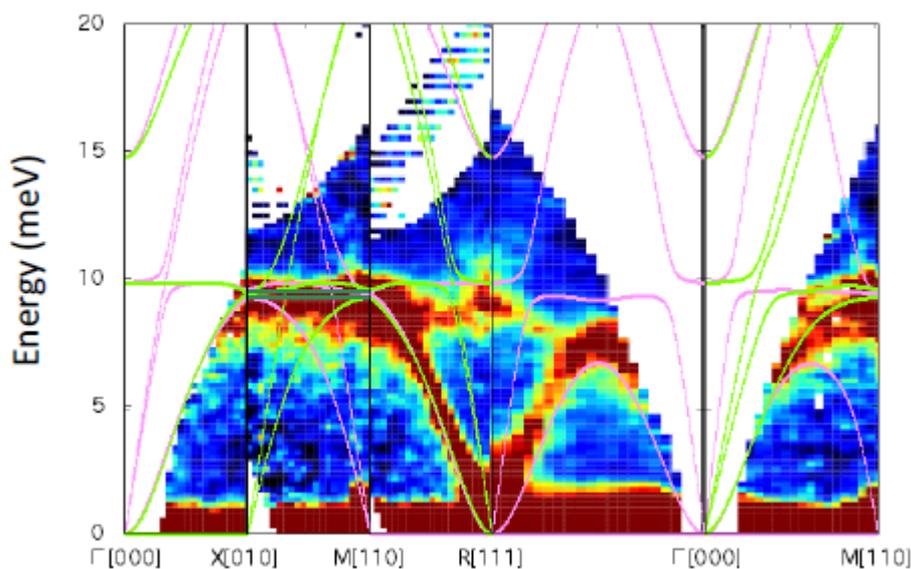


Figure 2. Experimental inelastic intensity observed at 5 K (counter map) and the corresponding calculated dispersion relations (green and pink lines) along the three high symmetry axes below 20 meV.