MLF Experimental Report	提出日 Date of Report 1 June, 2016
課題番号 Project No. 2015A0209 実験課題名 Title of experiment Observations of spin-orbit-lattice excitations in frustrated spinel MgV_2O_4 with orbital degree of freedom by single-crystal inelastic neutron scattering 実験責任者名 Name of principal investigator Keisuke Tomiyasu 所属 Affiliation Tohoku University	装置責任者 Name of responsible person Kenji Nakajima 装置名 Name of Instrument/（BL No.） AMATERAS (BL14) 実施日 Date of Experiment from 22 February to 1 March, 2016

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.

Spinel MgV_2O_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.

I. Introduction

Frustrated magnets are promising sources for exotic spin liquid/liquid-like states. Spinel AB_2O_4 provides the representative spin-frustrated systems, in which magnetic B ions form a corner-sharing tetrahedral lattice called a pyrochlore lattice. Insulating spinel MgV_2O_4 bears orbital degree of freedom [V^{3+}: d^2, spin $S = 1$, $(t_{2g})^2$] and its spin-orbital-coupled frustration has attracted much interest both theoretically and experimentally. Although the stoichiometric sample successively exhibits rather complex spin ordering and orbital-lattice ordering, a tiny amount of excess Mg holds the system disordered without any specific-heat anomaly at least down to 2 K [A. T. M. Nazmul Islam *et al*. (2012)].

Interestingly, recent ultrasound measurements found a novel macroscopic property in this disordered MgV_2O_4, that is, several elastic moduli significantly soften with decreasing the temperature from at least 150 K to 30 K [T. Watanabe *et al.* (2014)]. Because there is no phase transition, this lattice softening is not due to the well-studied precursor phenomena to structural transition. Further, the sample exhibits spin freezing at T_f = 13 K, hence, the softening to 30 K does not go towards the spin freezing. Instead, the ultrasound results are quantitatively analyzed as thermal activation effects of excitations around 5 meV to 15 meV, which are probably novel spin-orbit-lattice-coupled excitations.

Thus, by single-crystal inelastic neutron scattering, we observed the overall spin excitations up to ~ 30 meV, including this suggested energy range, in the disordered MgV_2O_4.

II. Experimental method

Standard inelastic neutron scattering experiments were performed using CCR on BL14 (AMATERAS).

III. Results & discussion

As shown in Figs. (a) and (b), the orientation-averaging spectrum structure extending along energy axis is observed. The scattering is gapless quasielastic at 15 K, which opens a finite gap ~2 meV at 4 K below T_f = 13 K. This excitation continues at least 30 meV (not shown).

Figure (c) shows the orientation-resolved $\boldsymbol{Q}$ dependence, sliced at constant energy. No energy dependence is detected within the entire measured energy range (not shown). The character of intensity pattern is well reproduced by that calculated for antiferromagnetic nearest-neighbor dimer or tetramer (tetrahedron), as shown in Fig. (d). These two models show almost the same patterns so cannot be distinguished. Further, these models are spatially rather isotropic, which is consistent with the fact that the ultrasound measurements detect no special symmetry for the excitations [T. Watanabe *et al.* (2014)].

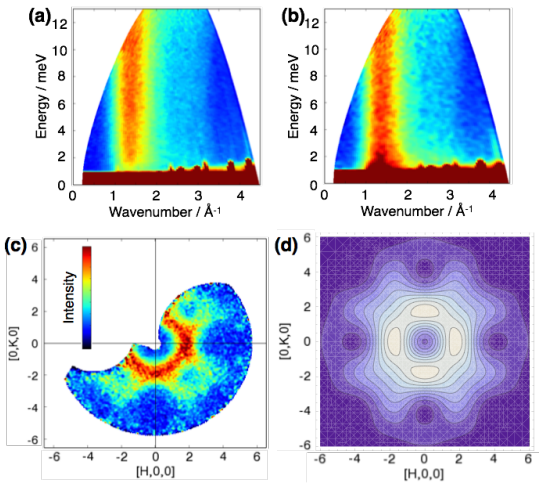

Figure. Experimental orientation-averaging data measured at 4 K (a) and 15 K (b). (c) Experimental and (d) calculated $\boldsymbol{Q}$ distribution map in the (h k 1/2) plane.

IV. Conclusions

We successfully observed and analyzed the molecular spin excitations in the disordered MgV_2O_4, which most probably correspond to the origin of the anomalous elastic property. Further details are under consideration.