Spin excitations of spin-1/2 frustrated square lattice magnet candidate $2VOSO_4 \cdot H_2SO_4 \cdot nH_2O$

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A spin nematic state is the exotic state in which spin rotation symmetry is broken but time-reversal symmetry is preserved [1]. The spin nematic state is expected in the frustrated square magnets with competing ferromagnetic nearest-neighbor (NN) interactions J_1 and nextnearest neighbor (NNN) antiferromagnetic interactions J_2 [2, 3]. Although several candidate compounds, such as BaCdVO₄(PO₄)₂ [4, 5] have been found so far, the model compounds often suffer from unoptimized parameters. It is still necessary to find better candidate compounds to reveal the nature of the spin nematic state.

We have found that Vanadyl sulfate hydrates, $2VOSO_4 \cdot H_2SO_4 \cdot nH_2O[6]$ can be also candidate compounds for the spin-1/2 frustrated square lattice magnet. This compound crystallizes in the space group of $P4_2/mnm$ at a room temperature. Exchange parameters were estimated as $J_1 \sim -1$ K and $J_2 \sim 5$ K from the fit to the high temperature series expansion. Magnetization and heat capacity measurements suggest the presence of the successive magnetic phase transitions at 1.9 and 2.3 K. In addition, neutron diffraction studies using the deuterated single crystalline sample have revealed the striped antiferromagnetic order below 1.9 K.

In this study, we have performed single inelastic neutron scattering crystalline experiments using a high-energy resolution triple-axis spectrometer HER. Totally 0.63 g of deuterated single crystalline samples were coaligned on two Aluminum plates with setting the HK0 plane as a horizontal scattering plane. The sample was coated by an amorphous fluoropolymer (CYTOP) and sealed in an aluminum can with He/N2 mixed exchange gas. The incident neutron energy of 3.6 meV was selected using pyrolytic graphite (PG) 002 reflections. A Be filter was installed in the upstream of analyzer to remove higher harmonic neutrons. The analyzer was set in the flat mode with the horizontal collimations of guide-open-80'-open. The sample was cooled down to 0.6 K using the closed cycle ³He refrigerator.

We performed several constant Q scans between 100 and 010 reflections at 0.7 K, which are shown in Figure 1. A clear single peak was observed between 0.2 0.8 0 and 0.8 0.2 0 positions. On the other hand, at the 100 position, the constant Q scan exhibits weakly decreasing behavior up to 1.2 meV, indicating that the peak is shifted to the elastic position and merged into the strong intensities induced from the Bragg reflections. The peak shift nearly from the elastic position to 1.0 meV should reflect the dispersion relation of magnons. The strong magnetic excitations extending up to 1.0 meV are consistent with those induced from the striped antiferromagnetic order with dominant antiferromagnetic $J_2 \sim 5$ K. Indeed, the peak shift well agrees with the dispersion from the expected exchange parameters, $4 \times 5 \text{ K/2} \sim 1.0$ meV. Careful analyses considering the resolution function and the sample mosaic are necessary to estimate J_1 of this compound.



Fig. 1. Constant Q scans of $2\text{VOSO}_4 \cdot D_2\text{SO}_4 \cdot nD_2\text{O}$ measured at 0.7 K. Some scans are shifted for clarity.

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