Successive phase transition in the spin-1/2 frustrated square lattice magnet $2VOSO_4 \cdot D_2SO_4 \cdot nD_2O$

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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 next-nearest neighbor (NNN) antiferromagnetic interactions J_2 [2, 3]. Although several candidate compounds, such as $BaCdVO_4(PO_4)_2$ [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, whereas the crystal symmetry is lowered below 280 K due to a structural phase transition. In magnetization and heat capacity measurements, anomaly was found at 1.9 and 2.4 K, suggesting the presence of the successive magnetic phase transitions. Neutron diffraction studies using the deuterated single crystalline sample have revealed the striped antiferromagnetic order below 1.9 K. On the other hand, a possible magnetic order at intermediate temperature is still not clear. Thus, we continued our single crystalline neutron diffraction experiments to reveal the magnetic structure at the intermediate temperature.

Single crystalline neutron diffraction experiments were performed by using a general-purpose neutron triple-axis spectrometer (GPTAS). A single crystalline sample with a size of 4 × 4 × 0.4 mm^3 was fixed on an Aluminum plate with setting the *HK*0 plane as a horizontal scattering plane. The sample was coated by an amorphous fluoropolymer (CYTOP) and sealed in an aluminum can with He exchange gas. The incident neutrons were monochromated to λ = 2.664 Å using the pyrolytic graphite (PG) 002 reflections. A PG filter was installed in the upstream of monochromator to remove higher harmonic neutrons. The spectrometer was operated in the double-axis mode with the horizontal collimations of 40’-80’-80’-open. The sample was cooled down to 0.7 K using the closed cycle ^3He refrigerator.

We performed several line scans to search for magnetic reflections. Fig. 1 shows a typical scan performed the along *H*00 and *HH*0 directions at 0.7 K (base), 2.1 K (intermediate), and 3.0 K (paramagnetic). Increase in intensities of the 100 reflection reflects the development of the striped antiferromagnetic order. The magnetic moments should be coupled ferromagnetically along the *c* direction at the base temperature. On the other hand, we could not find any differences in the intensities measured at 2.1 and 3.0 K at the *HK*0 position (-0.2 < *H* < 1.7, 0 < *K* < 1.1). This suggests that magnetic reflections in the intermediate phase are not present in the *HK*0 plane. If the magnetic moments are not coupled ferromagnetically along the *c* direction, the magnetic reflections can appear out of the *HK*0 plane. This possible scenario should be verified by neutron diffraction experiments on the *H*0*L* plane.

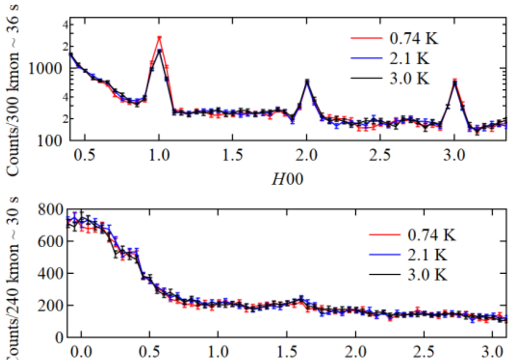

Fig. 1. Neutron diffraction patterns of $2VOSO_4 \cdot D_2SO_4 \cdot nD_2O$ measured at 0.7, 2.1, and 3.0 K.

[1] A. V. Chubukov, PRB **44**, 4693 (1991). [2] B. Schmidt *et al.*, PRB **76**, 125113 (2007). [3] N. Shannon *et al.*, PRL **96**, 027213 (2006). [4] R. Nath *et al.*, PRB **78**, 064422 (2008). [5] M. Skoulatos et al., PRB **100**, 014405 (2019). [6] P. M. Tachez and F. Theobald, Acta. Cryst. **B37**, 1978 (2008).