Neutron scattering study in magnetically ordered state in molecular quantum magnet

Shinichiro Asai, Toshiya Yoshida, Takatsugu Masuda
Institute for the Solid State Physics, The university of Tokyo

Geometrical frustration in magnets suppresses the spin ordering and induces novel magnetic states at low temperatures. The triangular lattice antiferromagnet with S =1/2 spins has attracted much attention since Anderson predicted the quantum spin liquid state [1]. In real compounds, the ion with S = 1/2 spin has the Jahn-Teller instability, which induces the lattice distortion at low temperature. The macroscopic degeneracy of the ground state is lifted owing to such a perturbation, which leads to the various types of magnetic order. One strategy to find a perfect triangular antiferromagnet is to focus on a magnet where molecular-like orbital is responsible for magnetism and the system is free from Jahn-Teller distortion.

$LiZn_2Mo_3O_8$ [2] and $Li_2InMo_3O_8$ [3] are the candidates for such a triangular lattice system. These compounds have the Mo_3 trimers which carry seven electrons and S = 1/2 spins in the molecular-like orbitals. The magnetic susceptibility and heat capacity measurements for $LiZn_2Mo_3O_8$ indicate a formation of a condensed valence-bond state below 100 K and no magnetic transition down to 0.1 K [2]. In contrast, those for $Li_2InMo_3O_8$ show the magnetic transition at 12.6 K [4]. 120 degree structure is expected as the magnetic structure of $Li_2InMo_3O_8$ from ^{7}Li NMR spectra at 4.2 K [4]. We performed neutron diffraction experiments in order to investigate the magnetic state of $Li_2InMo_3O_8$.

Powder neutron diffraction measurements were carried out at the high resolution powder diffractometer SPODI at FRM II. We used 10.6 g of polycrystalline sample synthesized by the solid state reaction. The temperature control was achieved by the closed-cycle refrigerator. The wavelength of 1.548 Å was obtained by the Ge511 monochromator. The neutron diffraction pattern for $Li_2InMo_3O_8$ at 20 K is shown in Fig. 1(a). The profile can be fit with the hexagonal structure with the space group $P6_3mc$. The obtained structural parameters are consistent with those reported in the previous study [3]. Figure 1(b) shows the diffraction patterns at 3.8 and 20 K. We found no magnetic peaks at 3.8 K. We simulate the magnetic diffraction pattern of 120 degree structure ignoring the effect of magnetic form factor. If the maximum intensity of the magnetic peak is within the statistical error of the background intensity, the magnitude of the moment is smaller than 0.31 μ_B. It is not consistent with the moment of 0.89 μ_B expected from the NMR spectra [4]. This may be owing to rather enhanced magnetic form factor of the Mo_3 trimer which suppresses the magnetic peaks more intensively than that of a magnetic ion does [5].

There is no difference between the profiles at 3.8 and 20 K within the experimental error, which suggests that the crystal structure of $Li_2InMo_3O_8$ keeps three rotation symmetry below the transition temperature.

[1] P. W. Anderson, Mat. Res. Bull. **8** (1973) 153. [2] J. P. Sheckelton *et al*., Nature materials **11** (2012) 493. [3] P. Gall *et al*., J. Solid State Chem. **208** (2013) 99. [4] Y. Haraguchi *et al*., Phys. Rev. B **92** (2015) 014409. [5] M. Mourigal *et al*., Phys. Rev. Lett. **112** (2014) 027202.

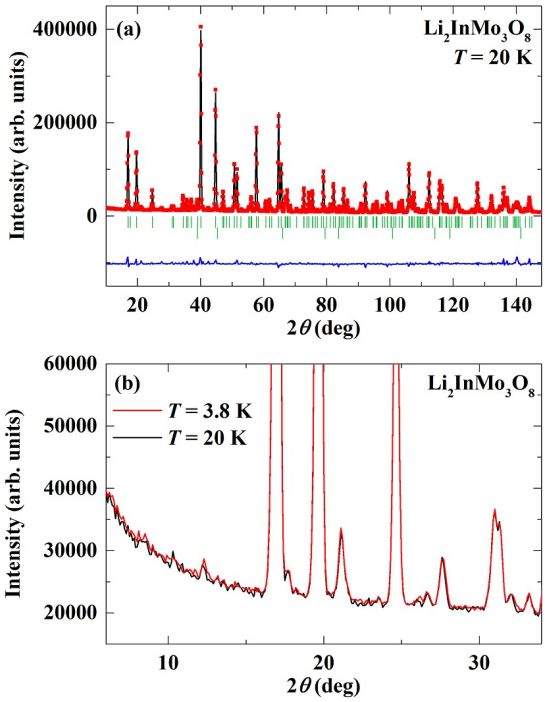

Fig. 1. (a). Rietveld refinement of neutron diffraction pattern for $Li_2InMo_3O_8$ at 20 K. (b) Neutron diffraction pattern for $Li_2InMo_3O_8$ at 3.8 and 20 K.