Magnetic ordering of isosceles-triangular lattice antiferromagnet $Mn(Nb_{1-x}Ta_x)_2O_6$

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Columbite compounds MnB_2O_6 (B=Nb, Ta) is one of model materials of Heisenberg antiferromagnets, where Mn magnetic ions form an isosceles triangular lattice in the *a*-*b* plane. Earlier neutron diffraction measurements [1,2] revealed that $Mn(Nb_{1-x}Ta_x)_2O_6$ exhibits a non-collinear magnetic structure, where a cant angle of Mn spins changes with x. However, there are many inconsistencies between reported magnetic structures, possibly due to deviations of compositions from the chemical formula and the difficulty of separating nuclear and magnetic reflections in powder diffraction data.

In this study, neutron diffraction experiments were performed for a single crystal of $MnNb_2O_6$ (x=0) using TOPAN (6G). The wavelength of incident neutrons was 2.475 Å. The single crystal with dimensions of approximately 2×2×2 mm^3, was mounted in a 1K refrigerator so as to access (H,K,0) or (H,0,L) scattering plane.

Figure 1 shows reciprocal lattice scans along the a^* direction at T=0.6, 4.0, and 6.0 K. In addition to nuclear reflections due to space group of *Pbcn*, magnetic reflections were found to develop below the Neel temperature of 4.3 K. Observed magnetic reflections could be classified into 2 groups;

(1) G-type for (H,K,L) with H+K=2n+1 and for (H,0,L) with L=2n+1, such as (1,0,1), (3,0,1), (5,0,1) reflections, and

(2) A-type for (H,K,L) with H+K=2n and for (H,0,L) with L=2n+1, such as (2,0,1), (4,0,1), (6,0,1) reflections.

As shown in Fig. 2, integrated intensities due to G-type and A-type magnetic structures exhibit different temperature dependence. This suggests a gradual change of orientation of Mn^{2+} spins with temperature and is consistent with previous work [2]. However, in our study, magnetic reflections which diminish at T_N, such as (3,0,0), (5,0,0), (0,0,1), were newly observed, indicating the appearance of a complicated magnetic structure, which is different from non-collinear structural model proposed previously [2].

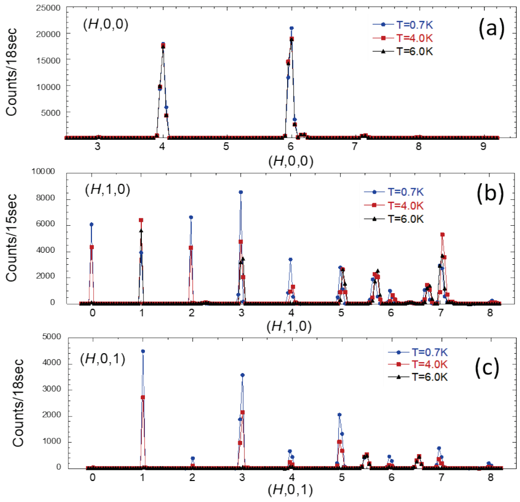

Fig. 1: Reciprocal lattice scans along the a^* direction at T=0.6, 4.0, and 6.0 K.

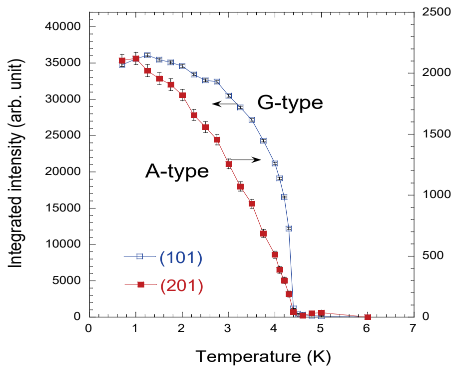

Fig. 2: Temperature dependence of integrated intensities at (1,0,1) and (2,0,1) Bragg points, originating from G-type and A-type magnetic structures, respectively.

[1] W. Scharf et al., Solid State Comm. vol.18 (1976) 249; V.S. Klein et al., Acta Cryst. vol.A32 (1976) 587.

[2] O.V. Nielsen et al., J. Phys. C: Solid State Phys. vol.9 (1976) 2401.