Magnetic diffuse scattering in pyrochlore antiferromagnet Na3Mn(CO3)2Cl

K. Nawa(A), D. Okuyama(A), M. Avdeev(B), and T. J. Sato(A)
(A)IMRAM, Tohoku Univ, (B)Bragg Institute, ANSTO

Spin systems with geometrical frustration have attracted much attention in the field of magnetism since they can exhibit unique magnetic structures, rich quantum phases, and critical phenomena. From this viewpoint, new compounds Na3T(CO3)2Cl (T = Mn, Co) should give an insight to approach the new quantum phases expected in pyrochlore antiferromagnets. This compound consists of TO_6 octahedra linked by carbonate ions (CO_3^{2-}), forming pyrochlore network of T^{2+} cations. For Na3Co(CO3)2Cl, previous macroscopic measurements and neutron scattering experiments [1] have revealed successive phase transitions in at T_a = 4.5 K and T_N = 1.5 K, described as a spin-glass like transition and all-in-all-out long-range magnetic order, respectively. However, the origin of the all-in-all-out magnetic order below the spin-glass temperature is still unknown. We believe that orbital degeneracy should be a key, since a coupling between the orbital degeneracy and a spin S = 3/2 for Co^{2+} (d^7) leads to a Kramers doublet of pseudo-spin 1/2 as a ground state. In fact, the temperature dependence of magnetic susceptibility is well reproduced by using high-temperature series expansions of spin 1/2 pyrocholore antiferromagnet.

To confirm our assumption, we prepared a new Mn-analogue, Na3Mn(CO3)2Cl. Its ground state should be different from Co-compound and may be intriguing because of the higher isotropy in magnetic interactions. In fact, the temperature dependence of magnetic susceptibility exhibits no clear anomaly down to 0.6 K, in contrast to Na3Co(CO3)2Cl. To investigate whether a magnetic order is present or not at low temperature, we performed neutron powder diffraction measurements using high-resolution powder diffraction spectrometer ECHIDNA at Bragg Institute, ANSTO. A dilution insert was used to reach down to 45 mK. Figure (a) shows diffraction patterns measured at 45 mK and 2.0 K, and difference of them. No additional magnetic Bragg peaks are present below 2.0 K, indicating the absence of a long-range magnetic order down to 45 mK. Instead, we find a diffuse scattering which develops below 50 K, as shown in Fig. (b). The bottom curve represents the intensity difference between 1.5 K and 200 K. A broad peak is clearly present at $Q \sim 0.85$ Å^{-1}, which is roughly close to $Q \sim 2\pi/d_{(110)}$ = 0.63 Å^{-1}, where $d_{(110)}$ = 10.04 Å corresponds to a doubled distance between nearest Mn-Mn atoms. This suggests that nearest neighbor antiferromagnetic correlations are dominant. Since the small difference in the spin correlations can be due to additional interactions such as dipolar-dipolar interactions, a further analysis for the diffuse scattering is necessary, which is in progress.

Our results reveal that Na3Mn(CO3)2Cl has a disordered ground state in contrast to Na3Co(CO3)2Cl which exhibits an all-in-all-out order. Since the former compound is a spin 5/2 system with isotropic magnetic interactions, the latter compound, which is pseudo spin 1/2 system, should have an anisotropy in magnetic interactions to exhibit the magnetic order. After analyzing the diffuse scattering patterns of Na3Mn(CO3)2Cl, reanalyzing those for Na3Co(CO3)2Cl using almost the same model except for single-ion and exchange anisotropy should reveal the origin of the unique magnetic transitions.

In summary, we conclude that a magnetic long-range order is absent down to 45 mK but antiferromagnetic short range order develops below 50 K for Na3Mn(CO3)2Cl. We are now analyz-

ing the diffuse scattering patterns to clarify magnetic interactions present in these unique pyrochlore compounds.

[1] Zhendong Fu et al., Phys. Rev. B, 87, 214406, (2013).

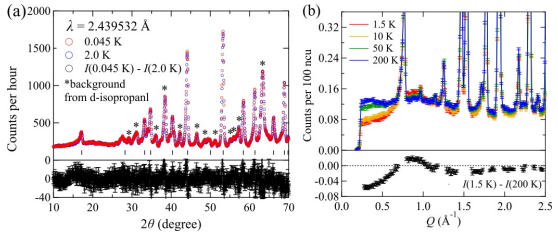

Fig. 1. (a) Neutron powder diffraction patterns measured at 0.045 K and 2.0 K. (b) Diffraction patterns measured at 1.5, 10, 50, and 200 K.