表題：擬スピン 1/2 パイロクロア反強磁性体 Na3Co(CO3)2Cl の磁気秩序

Magnetic order in the pseudo-spin-1/2 pyrochlore antiferromagnet Na3Co(CO3)2Cl

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Pyrochlore antiferromagnets exhibit rich magnetic structures, quantum phases and critical phenomena such as non-collinear spin structures, spin-liquids, spin-ices, and spin-glass transitions [1]. A lot of studies have been performed so far on rare-earth pyrochlores, where the ground state has zero or low degeneracy due to very strong spin-orbit coupling and crystal-field effects. The low-energy spin dynamics can often be reduced to that of a pseudo-spin-1/2 moment with anisotropic spin interactions and quantum effects, leading to nontrivial interesting phases such as a quantum spin ice in Yb2Ti2O7 [2]. From this viewpoint, the pyrochlore lattice formed by Co2+ ions (3d7) with octahedral coordinates should be also interesting. Na3Co(CO3)2Cl consists of CoO6 octahedra linked by carbonate ions, forming pyrochlore network of Co2+ cations. Previous macroscopic measurements and neutron scattering experiments [3] have revealed successive phase transitions in at Ta = 4.5 K and TN = 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. Since Mn-analogue Na3Mn(CO3)2Cl does not show any magnetic anomaly down to 0.05 K [4], we believe that orbital degeneracy leading to the single-ion anisotropy should be a key.

To confirm this expectation, we reinvestigated the magnetic structure below TN through powder neutron diffraction experiments using high-resolution powder diffractometer HB-2A at ORNL. A 3He insert was used to reach down to 0.25 K. Figure (a) and (b) show diffraction patterns measured at 0.25 K and 3 K, respectively. Clearly, additional magnetic Bragg peaks were observed below TN, indicating the occurrence of the long-range magnetic order. Among the decomposition of the irreducible representations for Co ions, Γ 1 representation well explains the whole powder diffraction pattern, supporting the all-in-all-out order indicated by the previous report [3]. The moment size is determined as 1.56(5) uB from the fit, slightly smaller than 2 uB expected from the g-factor determined from ESR experiments. In fact, a diffuse scattering developed at 3 K still persists at 0.25 K, indicating the presence of some sort of antiferromagnetic short range order well below TN (Figure (c)). Since the all-in-all-out order is confirmed from the present experiment, we are planning to examine the crystal field splitting from inelastic neutron scattering experiments as a next step, to reveal the origin of the all-in-all-out magnetic structure.

[1] J. Gardner et al., Rev. Mod. Phys. 82, 53 (2010).
[2] K. A. Ross et al., Phys. Rev. X 1, 021002 (2010).
[3] Z. Fu et al., Phys. Rev. B 87, 214406 (2013).
[4] K. Nawa et al., Phys. Rev. B 98, 144426 (2018).

使用施設：JRR-3M，装置：T1-3:HERMES
分野：Magnetism

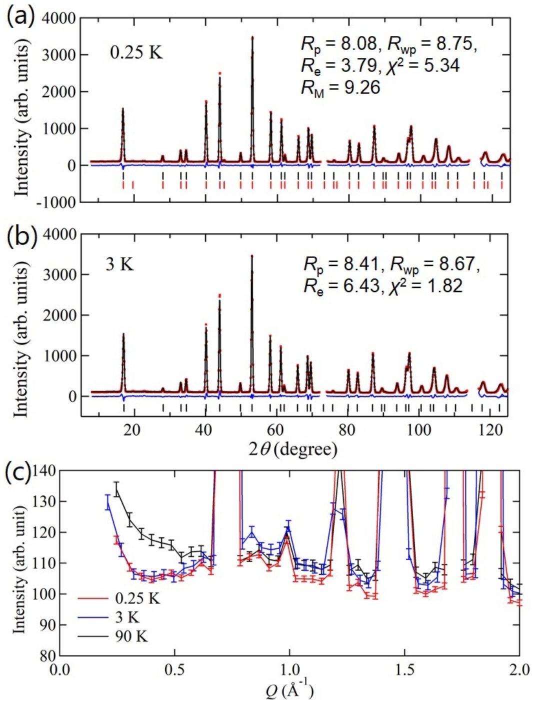

Fig. 1. (a, b) Neutron powder diffraction patterns measured at 0.25 K and 3 K. Red dots, black curves indicate observed, calculated intensities, respectively. Blue curves represent the intensity difference. (c) Q-dependence of the intensities at 0.25, 3, and 90 K.