

Magnetic Structures of (MX)LaNb₂O₇ (M=Cu, Mn...; X=Cl, Br)

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In recent years, much attention has been given to low-dimensional antiferromagnetic (AFM) quantum spin systems that have a ground state with no long-range magnetic order and an energy gap in the magnetic excitation spectrum. Among them, the $S = 1/2$ frustrated square lattice AFM system (J₁-J₂ model) is of special importance in light of Anderson's resonating valence bond (RVB) concept to explain high-T_c superconductivity in doped layered cuprates. Low-temperature topotactic strategies such as intercalation/deintercalation offer effective routes for the design and construction of new two-dimensional (2D) magnetic materials.

Topochemical ion exchange reaction has been employed to obtain a double-layered Dion-Jacobson phase such as (CuCl)LaNb₂O₇ starting from a nonmagnetic mother compound RbLaNb₂O₇ [1-4]. As shown in Fig. 1 (a), the structure consists of magnetic [CuCl] planes having an $S = 1/2$ square lattice arrangement that are widely separated by nonmagnetic double perovskite slabs. Thus it can be a very good candidate for the 2D quantum square lattice system. In 2004, we performed inelastic neutron scattering experiments on (CuCl)LaNb₂O₇ using the ISSP-PONTA triple-axis spectrometer installed at a 5G beam port of JRR-3M at the Japan Atomic Energy Research Institute. It has been revealed that this system indeed provides a new class of two-dimensional (2D) Heisenberg spin systems that has a spin-singlet ground state with a finite energy gap of 2.3 meV [5, 6]. Quite recently, we prepared isostructural compound (CuBr)LaNb₂O₇ using a method similar to that to obtain (CuCl)LaNb₂O₇. In spite of the fact that the lattice parameters are almost the same,

the magnetic properties of the copper bromine system is completely different from the spin-liquid state observed in the copper chlorine system. In contrast to the spin-gapped behavior in the Cl sample, the magnetic susceptibility of the Br sample shows the gapless behavior with an antiferromagnetic ordering at 31 K. In order to reveal the magnetic structure, we demonstrate the powder neutron diffraction profiles measured at 3 K and 45 K. We found two magnetic peaks centered at $2\theta = 15^\circ$ and 20° that can be indexed as $(1/2\ 0\ 1/2)$ and $(1/2\ 0\ 3/2)$, respectively, indicating the so-called collinear type (or stripe) ordering, where arrays of ferromagnetic chain along [100] align in an antiferromagnetic manner in the square lattice (see Fig. 1(a)). The collinear ordering is expected to occur when the magnitude of the next-nearest-neighbor exchange interaction is greater than that of the nearest-neighbor interaction. To date, only three examples have been reported to exhibit collinear ordering among $S = 1/2$ square lattice systems. Therefore, it would be interesting to examine the nature of the collinearly ordered state and to compare with the spin liquid state in the isostructural (CuCl)LaNb₂O₇.

To further understand the nature of the unusual ground states observed in (CuCl)LaNb₂O₇ and (CuBr)LaNb₂O₇, it would be quite interesting to compare these compounds with other related materials. Thus, in this study, we performed powder neutron diffraction experiments on the isostructural (MCl)LaNb₂O₇ (M=Co, Cr, Mn), using the IMR-HERMES diffractometer (T1-3). Neutrons with a wavelength of 1.81386(7) Å were obtained by the 331 reflection of the Ge monochro-

mator, and the 12'-blank-sample-18' collimation was employed. A powder sample of 4 g was put into a vanadium cylinder. Figure 1(b) demonstrates the neutron diffraction profiles of (CoCl)LaNb₂O₇ obtained at 8 K and 100 K. The lower temperature data have additional magnetic reflections characterized by the propagation vector (π , 0, π), indicating the collinear type of magnetic ordering. The magnetic ordering temperature is obtained to be 55K by measuring the intensity of (1/2, 0, 1/2). The manganese analogue also exhibits the collinear order but at slightly higher temperature, while the neutron diffraction profile of the chromium analogue may indicate incommensurate magnetic structure at low temperatures. This work has been supported by Grants-in-Aid for Young Scientists (A) (No. 17684018) and Science Research on Priority Area (No. 16076210) from MEXT of Japan, and by the 21st Century COE Program, Kyoto University Alliance for Chemistry.

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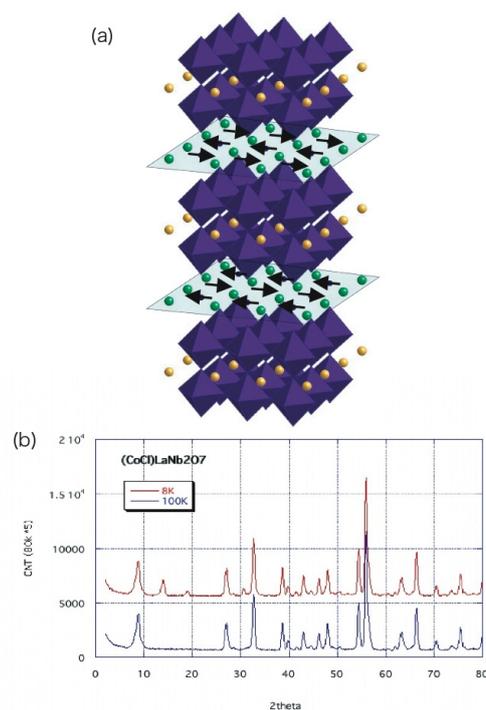


Fig. 1. (a) Crystal and magnetic structure of (MX)LaNb₂O₇. (b) Neutron diffraction patterns for (CoCl)LaNb₂O₇ measured at 8 K and 100 K, demonstrating the collinear (π , 0) type of magnetic long range ordering.