Magnetic states in Layered metal oxyhalides A_2MO_3X and $A_2MO_2X_2$

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A magnetic phase diagram in a square-lattice antiferromagnet is one of the hot topics in the field of magnetism. In case that diagonal next-nearest-neighbor interaction J_2 is antiferromagnetic and the nearest-neighbor interaction J_1 is ferromagnetic, J_2 competes with J_1 and, thus, the frustration induces a novel phase called the spin-nematic phase in the J_1 - J_2 phase diagram.[1)] (CuCl)$LaNb_2O_7$ was considered as a candidate for the nematic state[2)] but a recent neutron study revealed that the ground state is a conventional spin-singlet state.[3)] Layered metal oxyhalides A_2MO_3X and $A_2MO_2X_2$ (A = Ca, Sr, M = Mn, Fe, Co, Ni, Cu, X = F. Cl) belong to a new family of the square-lattice antiferromagnet.[4),5)] They could be a model for the frustrated square-lattice antiferromagnets. We have synthesized some of the compounds and measured the bulk properties. Among them Sr_2NiO_3Cl, $Sr_2NiO_2Cl_2$ and Sr_2MnO_3F are particularly interesting. The space groups are tetragonal $I4/mmm$ for $Sr_2NiO_2Cl_2$ and Sr_2MnO_3F, and $P4/nmm$ for Sr_2NiO_3Cl. We performed neutron powder diffraction (NPD) experiments and Rietveld analyses to determine the magnetic structures. The wave length of the neutron beam was $\lambda = 2.4395(3)$ Å throughout the experiments. We have used the software FullProf for the Rietveld analyses.

The diffraction profiles of Sr_2MnO_3F at 3 K and 180 K are shown in Fig. 1 (a). The order of O/F site was not observed. The refined cell parameters are $a = b = 3.78751(6)$, $c = 13.26733(24)$ at 180 K and $a = b = 3.78137(1)$, $c = 13.26237(27)$ at 3 K. In addition to nuclear Bragg peaks at 180 K, new peaks are observed at $T = 3$ K. Since an anomaly was observed in the magnetic susceptibility at $T \sim 120$ K in our preliminary study, the origin of these peaks are magnetic long-range order. The propagation vector is identified as $q = (1/2, 1/2, 0)$. We performed representation analysis and obtained three possible models. We performed Rietveld analysis and obtained the collinear magnetic structure as shown in Fig. 1(b). The magnitude of the magnetic moment is estimated as $3.04(4)\mu_B$. Since the Mn^{3+} ions surrounded in O^{2-} octahedra are responsible for the magnetism, the size of the spin in the electric ground state is $S = 2$ in case of high spin state. The spin moment in this compound is strongly suppressed from the full moment of 4 μ_B probably because of the low dimensionality of the spin system.

The NPD profiles of $Sr_2NiO_2Cl_2$ are shown in Fig. 1(c). The cell parameters are refined as $a = b = 4.03529(4)$, $c = 15.02470(26)$ for 270 K and $a = b = 4.03274(5)$, $c = 14.92936(31)$ for 3 K. Magnetic Bragg peaks are observed at the low temperature. The propagation vector is $q = (1/2, 1/2, 0)$ and the obtained magnetic structure is similar to Sr_2MnO_3F. The estimated magnitude of the magnetic moment is $1.20(15)\mu_B$ and it is strongly suppressed from the full moment of Ni^{2+} ion $2.0\mu_B$.

We performed NPD experiments on Sr_2NiO_3Cl at $T = 3$ K (21 hours), 40 K (3hours), 80 K (7hours), 120 K (4.5hours), 190 K (3hours) and 270 K (3hours). No lattice distortion was observed down to $T = 3$ K and the refined cell parameters are $a = b = 3.83388(3), c = 14.34838(20)$ at 80 K and $a = b = 3.83385(3), c = 14.34465(24)$ at 3 K. We subtract the NPD at 80 K from that at 3 K and the subtraction is shown in Fig. 1 (d). No peak is identified and this indicates the absence of the magnetic long-range order at 3 K. We think that the magnetic order is disturbed by a spin frustration and the magnetic state at low temperature may be a nematic state predicted in the frustrated J_1 - J_2 antiferromagnets.

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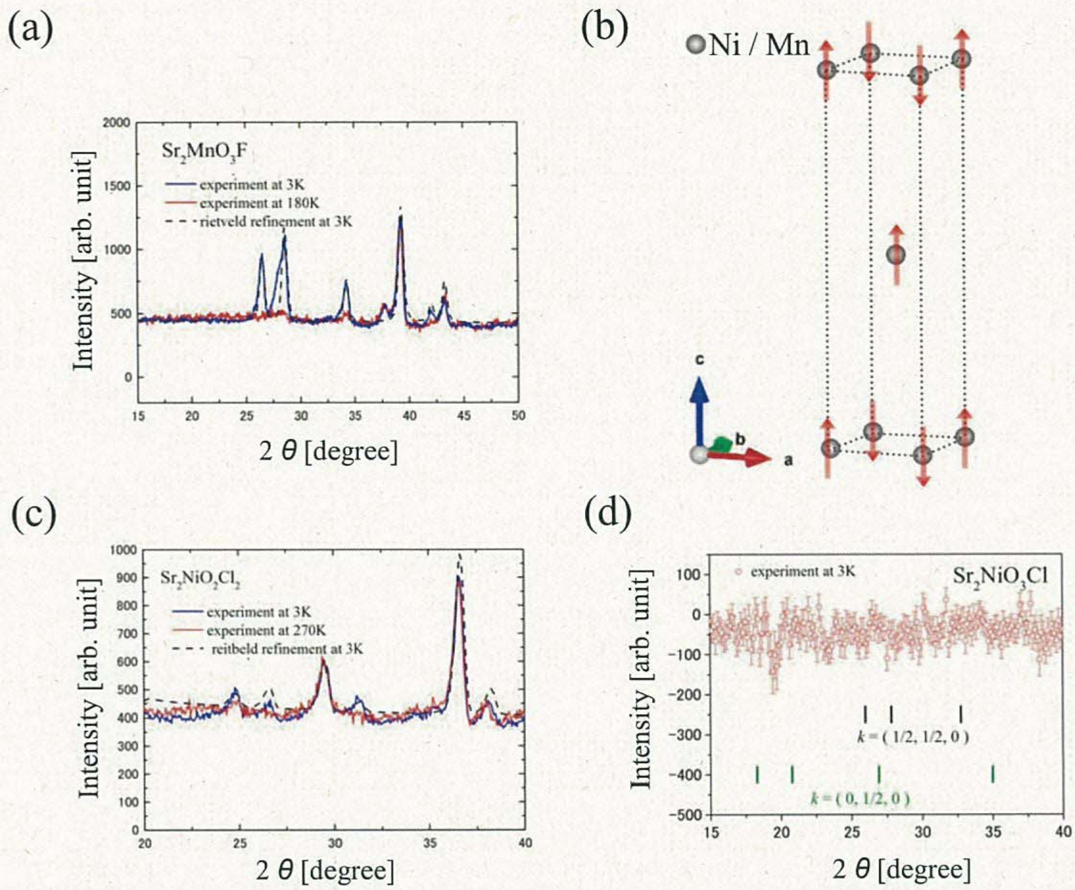

Fig. 1. (a) Neutron diffraction profiles of Sr_2MnO_3F. (b) Magnetic structure of Sr_2MnO_3F. (c) Neutron diffraction profiles of $Sr_2NiO_2Cl_2$. (d) Subtracted neutron diffraction profile of Sr_2NiO_3Cl.