Novel magnetic state in a series of new square-lattice magnets

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Two-dimensional square-lattice antiferromagnet (AFM) is a well-known playground for condensed matter science. Particularly those of K_2NiF_4 type structure have been extensively studied in the field of high-T_c superconductivity. In magnetism, on the other hand, the physics of the plain AFM had made considerable progress in the past several decade. Current interest is a novel magnetic state induced by additional interactions. In case that diagonal antiferromagnetic interaction J_2 is not negligible and the nearest neighbor interaction J_1 is ferromagnetic, J_2 compete with J_1 and the frustration gives rise to a nematic state in the vicinity of Neel Phase in the J_1 - J_2 phase diagram [1]. (CuCl)$LaNb_2O_7$ had been considered as a candidate [2] but recent neutron study revealed coupled Shastry-Sutherland quantum spin singlet [3]. Further searches for new model compounds is very important.

Layered metal oxyhalides A_2MO_3X and $A_2MO_2X_2$ (A = Ca, Sr, M = Mn, Fe, Co, Ni, Cu, X = F. Cl) are members of a new family of 2D square lattice antiferromagnets [4, 5]. We have synthesized several new compounds and measured several bulk properties. Since Sr_2NiO_3Cl, $Sr_2NiO_2Cl_2$ and Sr_2MnO_3F are more interesting, we performed neutron powder diffraction (NPD) measurements and Rietveld refinement to determine the crystallographic and magnetic structures. The wave length of neutron beam λ was the same on the all experiments and $\lambda = 2.4395(3)$ Å. We used FullProf software for the Rietveld refinements.

The diffraction profiles of Sr_2MnO_3F at 3 K and 180 K are shown in Fig. 1(a). The space group is $I4/mmm$. The obvious evidence of O/F site ordering was not observed. The cell parameters at 180 K and 3 K are $a = b = 3.78751(6)$ Å, $c = 13.26733(24)$ Å and $a = b = 3.78137(1)$ Å, $c = 13.26237(27)$ Å. Magnetic bragg peaks are clearly observed at low temperatures ($\leq$ 100 K). The propagation vector was identified as $q = (1/2, 1/2, 0)$. Combination of the representation analysis and Rietveld refinement leads to the collinear magnetic structure shown in Fig. 1(b). The direction of magnetic moment is parallel to the c^* axis and its magnitude is 3.04(4) μ_B. The result means that Mn^{3+} ion has a high spin state and the magnetic moment is strongly suppressed.

The diffraction profiles of $Sr_2NiO_2Cl_2$ at $T = 3$ K and 270 K are shown in Fig. 1(c). The space group is $I4/mmm$. The cell parameters at $T = 270$ K and 3 K are decided as $a = b = 4.03529(4)$ Å, $c = 15.02470(26)$ Å and $a = b = 4.03274(5)$ Å, $c = 14.92936(31)$ Å. The magnetic bragg peaks were observed at $2\theta = 24.70°$, 26.47° and 31.23° at $T = 3$ K. The propagation vector is $(1/2, 1/2, 0)$. The determined magnetic structure is the same as that of Sr_2MnO_3F as shown in Fig. 1(b). The magnitude of the moment is 1.20(15) μ_B. Similar to Sr_2MnO_3F the magnetic moment is strongly suppressed.

We performed NPD experiments at several temperatures on Sr_2NiO_3Cl. The obtained cell parameters at 80 K and 3 K are $a = b = 3.83388(3)$ Å, $c = 14.34838(20)$ Å and $a = b = 3.83385(3)$ Å, $c = 14.34465(24)$ Å. The profile is consistent with the space group $P4/nmm$ which has O/Cl site ordering, and no lattice distortion was observed down to $T = 3$ K. No magnetic peak is identified, which indicates the absence of the magnetic long-range order even at 3K (Fig. 1(d)).

Reference

[1] N.Shannon *et al.*, Phys. Rev. Lett. **96**, 027213 (2006) [2] H. Kageyama *et al.*, J. Phys. Soc. Jpn. **74**, 1702 (2005) [3] C. Tassel

et al., Phys. Rev. Lett. **105**, 167205 (2005) [4] C. S. Knee *et al.*, Phys. Rev. B **68**, 174407 (2003) [5] Y. Tsujimoto *et al.*, Inorg. Chem. **51**, 4802 (2012)

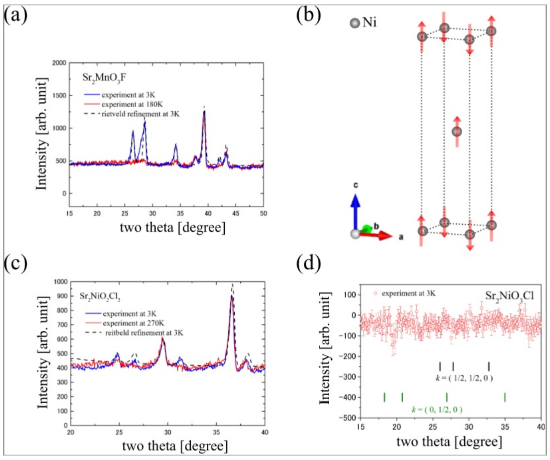

Fig. 1. (a) profiles of Sr_2MnO_3F (b) The magnetic structures of Sr_2MnO_3F and $Sr_2NiO_2Cl_2$ (c) profiles of $Sr_2NiO_2Cl_2$ (d) The profile of Sr_2NiO_3Cl at 80 K subtracted from that at 3K