Magnetic Structure in the Multiferroic Phase of Frustrated Magnet CuFe1-xAlxO2

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Geometrically frustrated triangular lattice antiferromagnet CuFeO2 has been extensively studied for last fifteen years. Recently, Kimura and co-workers has discovered the spontaneous electric polarization in the low-temperature field-induced phase of CuFeO2, and argued that CuFeO2 provides an opportunity to study a new class of multiferroic materials.[1] To understand the microscopic mechanism of the multiferroic nature, detailed magnetic structure in the ferroelectric(FE) phase is indispensable. Quite recently, we performed neutron diffraction measurements with a horizontal field cryomagnet at HMI, and revealed that the magnetic structure in the FE phase is a proper helical structure propagating along \(<110>\) axis.[2,3] This measurements is, however, restricted to the (hhl) zone, in which the proper helical structure cannot be distinguished from the sinusoidal structure with the moments canted by 45[deg] from the c axis toward \(<1-10>\) direction.[4] Thus, the information beyond the (hhl) zone is indispensable to confirm the proper helical structure.

Fortunately, the small magnetic reflections corresponding to the magnetic ordering of the FE state were found to coexist with the four-sublattice(up-up-down-down) magnetic state in the ground state of the slightly diluted sample CuFe1-xAlxO2(x=0.012), owing to a slight macroscopic inhomogeneity of the Al-concentration. We thus surveyed the three-dimensional hkl-dependence of the magnetic structure factor of the FE state in zero field, using the x=0.012 sample and four-circle diffractometer FONDER(T2-2) installed at JRR-3M. The incident neutron wavelength is 1.24[A] and the closed-cycle He-gas refrigerator is used to provide access to the temperature down to T=2.5K.

From observed magnetic structure factors, we estimated the Spin Orientation Factors (SOF), and found that the hkl-variation of observed SOF is well explained by the proper helical model, even beyond (hhl) zone, as shown in Figs. 1(b)-(e).

A naive application of the theory for ferroelectricity in noncollinear magnets[5] to the proper helical structure does not lead to a finite uniform electric polarization. Nevertheless, the present results imply that a spin noncollinearlity is relevant to the multiferroic nature in CuFe1-xAlxO2.

Fig. 1. (a) Typical diffraction profile at T=2.5K. (b)-(e) Index l-variation of and the SOFs. The solid, dotted and dashed lines denote the calculated values for a proper helical, +45[deg]-canted sinusoidal and -45[deg]-canted sinusoidal structure, respectively.