Magnetic structure of multiferroic $DyMnO_3$ studied by polarized neutron diffraction

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Recently, perovskite manganites have been intensively studied from the view points of multiferroics properties [1]. In typical multiferroics material $TbMnO_3$ and $DyMnO_3$, polarization flop phenomena are observed under the magnetic field [1]. Recent theoretical studies [2, 3] suggest that the cycloidal magnetic structure of Mn ion plays an important role to the ferroelectric state. In Ref. [2], ferroelectric polarization of multiferroics $TbMnO_3$ and $DyMnO_3$ can be expressed by cross product between the magnetic propagation vector and magnetic chirality from cycloidal structure as $P = \alpha\ k \times (S_i \times S_j)$. On the basis of this theory, the origin of polarization flop phenomena from $P//c$ to $P//a$ can be understood as being due to the magnetic structural phase transition from *bc*-cycloidal magnetic structure to *ab*-cycloidal magnetic structure. For ensuring this theoretical prediction, the magnetic structure of $P//c$ phase at zero filed and $P//a$ phase under hight magnetic fields should be clarified. Therefore, we utilized polarized neutron diffraction method and tried to decide the precise magnetic structure in both zero and high magnetic fields.

As the target material, we chose $DyMnO_3$, which shows the polarization flop phenomenon at low magnetic filed (~2 Tesla). Neutron diffraction experiments were performed on TOPAN triple-axis spectrometer. Neutron beam with an energy 80 meV was used. A single crystal of $DyMnO_3$ was mounted in longitudinal magnetic filed-type superconducting magnet (Endoh-magnet) so that the [1 0 0]-[0 0 1] direction on *Pbnm* setting were parallel to the scattering plane.

First, we tried to measure the eight magnetic reciprocal lattice points marked with red circles in Fig. 1 with unpolarized neutron diffraction mode for charactering the magnetic properties at low temperature of $DyMnO_3$. In Fig. 1, the peak profiles of magnetic reflections at (0 k 5), (0 k 6), (1 k 5), (1 k 6), (5 k 0), (6 k 0), (4 k 1), (5 k 1) positions are shown. Black triangle, red triangle, and blue circle indicate the peak profiles at T=15 K, 8 K, and 6.5 K near the ferroelectric ordering temperature T_C=18 K and the magnetic ordering temperature of Dy moment T_{N-Dy}=6.5 K, respectively. In the data at 8 K (6.5 K$\leq T \leq$18 K), only (0 0.38 5) and (4 0.38 1) reflections were observed, in which the magnetic moment of Mn ion is mainly involved. In contrast, in the data at 3 K ($T \leq$6.5 K), additional (0 1/2 5) and (5 1/2 0) were observed. These experimental results are consistent with the data of Ref. [4].

In order to decide the magnetic structures of Mn moment at both zero and high magnetic fields, we utilized polarized neutron diffraction method. For the limitation of the experimental configuration, we measured only (0 0.38 5) magnetic reflection. In this case, we can mainly observe the magnetic reflection from *a*-axis (*b*-axis) magnetic component at spin flip (non-filp) process, respectively. In Fig. 1, at 0.5 Tesla (near zero field $P//c$ phase), only the magnetic reflection from non-spin flip process was observed. This result strongly suggests the *bc*-cycloidal magnetic structure of Mn moment, which is consistent with the data of Ref. [4]. On the other hand, at 3.5 Tesla ($P//a$ phase), the small magnetic reflections from both non-spin flip and spin flip processes were observed with slightly poor statistics. From this result, we concluded that the *ab*-cycloidal magnetic structure of Mn moment is realized in $P//a$ phase at high magnetic fields.

In summary, we determined roughly the magnetic structure of Mn moment of multiferroic $DyMnO_3$ in both $P//c$ phase at low magnetic fields and $P//a$ phase at high magnetic fields. These magnetic structures support the theoretical prediction that the polarization flop phenomenon is due to the change of the cycloidal spin plane from *bc*-plane to *ab*-plane.

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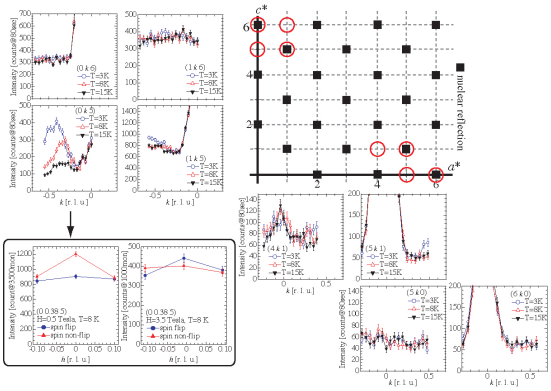

Fig. 1.