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Superlattice Diffraction in Multiferroic Manganite $DyMn_2O_5$

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Introduction

A family of RMn_2O_5 (R: rare earth) is known as the magnetic ferroelectrics which undergo successive ferroelectric transitions below the antiferromagnetic transition temperature T_N=39-45 K [1]. Very recently, it has been reported that RMn_2O_5 shows a gigantic magneto-electric effect related to the magnetic improper ferroelectricity. To investigate the correlation among its magnetism, structure, and electric property, we have performed the synchrotron x-ray diffraction study using a single crystals of $DyMn_2O_5$ (several mm in size) grown with use of PbO-PbF_2 flux [2]. The measurements were performed by using a Huber six-axis diffractometer at BL-4C. The incident x-ray was monochromated at 13 keV by utilizing a Si (111) double-crystal monochromator and focused on a (001) surface of a single crystal mounted in a closed-cycle He refrigerator. To reduce the contamination of luminous X-ray, a pyrographite (002) monochromator was installed in front of the scintillation counter.

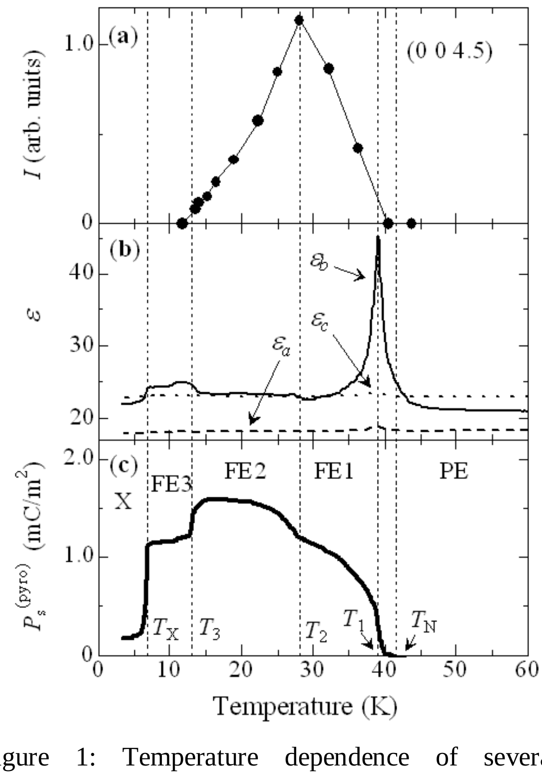

Figure 1: Temperature dependence of several quantities in $DyMn_2O_5$; (a) the integrated intensity (I) of x-ray superlattice peak at (0 0 4.5), (b) the dielectric constants for 1 kHz, and (c) the polarization ($\boldsymbol{P}_s^{(pyro)}$) along b obtained by the measurement of pyroelectric current in an applied field of 200 kV/m. All the temperature scans were done in the cooling run.

Results and Discussions

The x-ray superlattice reflection at (0 0 4.5) is observed to increase in lowering temperature from T_1, but tends to decrease in intensity below 28 K (=T_2) and then finally disappears around 13 K (=T_3), as shown in Fig. 1(a). The emergence of such a C lattice modulation is likely to correspond to the magnetic modulation with the propagation vector (1/2 0 1/4) reported in the temperature range between 18 K and 44 K by Wilkinson *et al.* [1]. As observed in the case of the ferroelectric perovskite-type $RMnO_3$ [3], the strong exchange-striction effect inherent to the Mn-O network produces the lattice modulation with twice of the modulation vector of the spin modulation. In analogy to the improper ferroelectrics like Rb_2ZnCl_4 and related compounds [4,5] as well as to perovskite $RMnO_3$, the IC to C transition of lattice modulation may produce the parasitic ferroelectricity also in this compound. The respective ferroelectric phases and their variation appear to be closely related to the successive emergence of the spin order with different propagation vectors (k_s), and the part of the lattice modulation caused by the commensurate (k_s=(1/2 0 1/4)) spin order can be seen as the superlattice peaks at $2k_s$ in the FE1 and FE2 phases. However, we could not observe (δ 0 9/2) superlattice in the FE3 phase where the magnetic modulation vector is reported to be incommensurate (δ/2 0 1/4) [1], which implies that the displacement of Mn and O may change in direction and become confined in the *ab* plane.

References

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