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Superlattice Modulation in Magnetoelectric Perovskite Manganites RMnO$_3$

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Introduction

Research on coupling between ferroelectric (FE) and magnetic orders has revived after Kimura *et al.* reported magnetic-field-induced flop of electric polarization P in TbMnO$_3$.[1] Evidence has been increasing that this unique ME effect is inherently connected to the Mn-spin ordering structure. There are two possible mechanisms of spin-order-driven ferroelectricity. It was suggested that the ferroelectricity in RMnO$_3$ can be attributed to the *symmetric* exchange striction at the earlier stage, due to the locking of the antiferromagnetic (AFM) wavenumber q_{mag} at the FE Curie temperature T$_C$. The superexchange interaction via oxygen ion between two Mn ions can be modified with a Mn-O-Mn bond angle.[2] Conversely, a Mn-O-Mn bond angle should be more or less affected by the spin arrangement. In RMnO$_3$, Mn-O-Mn bonds are not straight due to rotation and tilt of the MnO6 octahedra. Therefore, the Mn-O-Mn-O-… bonding along <110> forms a zigzag chain. Below the AFM transition temperature T$_N$, each Mn-O-Mn bond angle may be further influenced by the alignment of Mn moments. In the original zigzag chain, the oxygen ions between up and down spins move in the same direction when the magnetic modulation vector is represented as m/n, where m and n are odd and even integers, respectively.

As an alternative mechanism of the spin-order-driven ferroelectricity, it has recently been pointed out that transverse spiral spin ordering can induce FE polarization regardless of the commensurability between spin modulation and the lattice.[3] This mechanism is closely related to the *antisymmetric* superexchange or the so-called Dzialoshinski-Moriya (DM) interaction.[4] A bending Mn-O-Mn bond lacking an inversion center favors a canted spin arrangement. This antisymmetric superexchange term is linear to the spin-orbit coupling term, and is expressed as D·(S_i×S_j), where D is a constant vector. By considering the inverse effect, the stable position of the oxygen ion between two Mn ions can be modified using the vector product of the two magnetic moments, S_i×S_j. Since the vector product of any two neighboring Mn moments is constant in case of transverse spiral ordering, all the oxygen ions are displaced in the same direction.

Results

Superlattice reflections with a modulation vector (0 $2q_{mag}$ 0) emerge in AFM phases of perovskite RMnO$_3$ compounds with a spin modulation of (0 q_{mag} 1). Figure1 shows the temperature dependence of superlattice wavenumber $2q_{mag}$ in various RMnO$_3$ compounds. One can note a general tendency that a compound with larger rare earth element shows a modulation with smaller q_{mag}. Though the wavenumber q_{mag} varies above T_C in many compounds, lock-in transition is absence in some (Tb,Dy) derivatives. We hence doubt that the FE transition could be ascribed to the lock-in phenomenon of the AFM modulation vector.

We could determine the x value in Tb$_{1-x}$Dy$_x$MnO$_3$ with q_{mag}=1/3 from the figure. The compound with q_{mag} of 1/3 enabled us to perform a magnetic structure analysis without assuming any model. We performed a neutron diffraction measurement using FONDER at JRR-3 and proved that the transverse spiral AFM ordering causes the FE polarization.[5]

Fig. 1: Temperature dependence of antiferromagnetic modulation wavenumber q_{mag} in perovskite-type RMnO$_3$ compounds with various rare earth R. Arrows indicate ferroelectric transition temperature.

References
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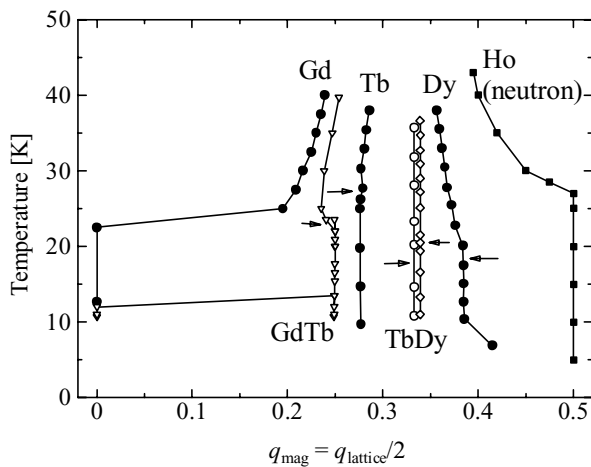
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