Spin-Lattice Coupling in $MnWO_4$, a Multiferroic Compound

A synchrotron X-ray study of the magnetic ferroelectric $MnWO_4$ in a magnetic field has been conducted. Superlattice modulation with a propagation vector twice as large as the magnetic propagation vector was observed in three magnetic phases in the absence of a magnetic field. The intensity of the superlattice reflection showed a sudden change upon the spiral-to-collinear magnetic transition, indicative of the usefulness of synchrotron X-ray for investigating the magnetic structure of the multiferroic compounds. A superlattice measurement in a magnetic field confirmed that the magnetic-field induced ferroelectric transition in $MnWO_4$ at low temperature is accompanied by the commensurate-collinear to incommensurate-spiral magnetic phase transition.

Ferroelectricity has been reported in several frustrated magnets. Such compounds often undergo successive magnetic transitions, and ferroelectricity is usually observed only in spiral magnetic phases. Although the X-ray diffraction technique cannot be used to determine completely unknown magnetic structures, it can be used to provide some useful information on long-wavelength spin alignment, and as a consequence, on the electric properties of the compounds [1].

$MnWO_4$ is a multiferroic material, in which ferroelectricity is induced by the cycloidal spin structure [2] Neutron diffraction results [3] revealed the AF1 ($T < T_1$), AF2 ($T_1 < T < T_2$) and AF3 ($T_2 < T < T_N$) phases, a commensurate collinear antiferromagnetic phase, an incommensurate cycloidal-spiral phase, and an incommensurate sinusoidal antiferromagnetic phase, respectively, as illustrated in Fig. 1. Ferroelectric polarization is observed along the monoclinic *b*-axis only in the spiral AF2 phase. The application of a magnetic field along the *a*- or *c*-axis in the AF1 phase can induce a ferroelectric transition. We have investigated the coupling between the lattice and the spin order in $MnWO_4$ using synchrotron-X-ray diffraction [3].

A single crystal boule of $MnWO_4$ was grown by a floating zone method. Off-resonant single crystal X-ray diffraction measurements were performed on BL-3A. A sample with cleaved (010) surfaces was cut from the boule and mounted in a He cryostat equipped with an 8 Tesla superconducting magnet. Superlattice reflections around (h −5 l) with h~1/2, l~0 were studied using an X-ray beam of photon energy 14 keV at several magnetic fields and temperatures.

An intense commensurate superlattice peak is discernible in the AF1 phase, and an incommensurate peak in the AF2 and the AF3 phases. The emergence of the superlattice modulation corresponds to the magnetic modulation, since the magnitude of the lattice propagation vector ($\boldsymbol{q}^l$) of each phase is almost twice the magnitude of the magnetic propagation vector ($\boldsymbol{q}^m$) obtained from neutron diffraction measurements [3]. The temperature dependence of the $\boldsymbol{q}^l$ value is shown in Figs. 2(a) and (b). The a*- and c*- components of the lattice propagation vector, q_x^l and q_z^l, continuously shift from the AF3- to the AF2- phase around T_2, whereas q_x^l and q_z^l discontinuously change to commensurate values, q_x^l =−0.5 and q_z^l = 1, around the incommensurate (AF2)-commensurate (AF1) transition temperature (T_1). The integrated intensity of the superlattice peak is plotted as a function of temperature in Fig. 2(c). The incommensurate peak gradually grows in intensity at temperatures below T_N until the development of the superlattice peak intensity is suppressed just below T_2. The commensurate peak in AF1 is an order of magnitude larger than that in AF2, indicative of the exchange striction mechanism of the superlattice.

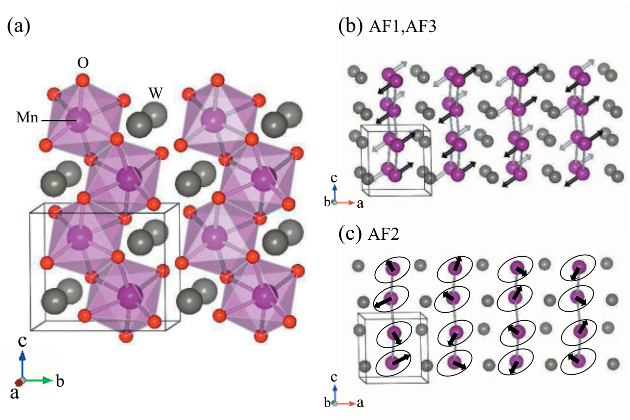

Figure 1
(a) Crystal structure of $MnWO_4$ viewed along the *a*-axis: each Mn atom is surrounded by an oxygen octahedron. W atoms separate zigzag chains of Mn atoms. (b) Collinear magnetic structure in AF1 and AF3: magnetic moments lie in the ac plane and canted to the *a*-axis by about 35 degrees. (c) Spiral spin structure in AF2: basal plane of spiral is inclined to the *ab* plane. Reproduced from Ref. [2].

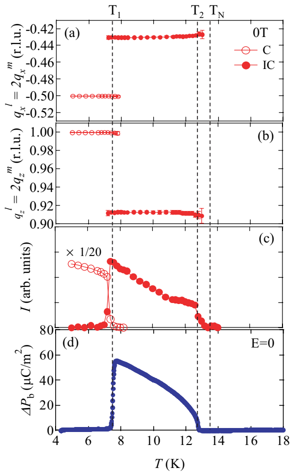

Figure 2
Temperature dependence of [(a) and (b)] the magnitudes of the lattice propagation vectors q_x^l and q_z^i, (c) integrated intensities of the incommensurate (closed circles), commensurate (open circles) superlattice peak, and (d) electric polarization along the b axis at 0T. Reproduced from Ref. [4].

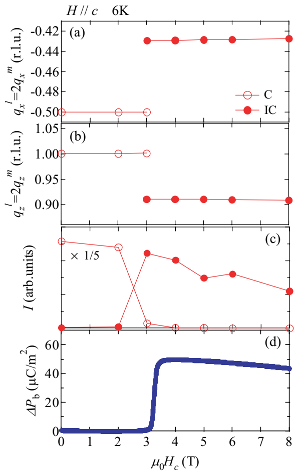

Figure 3
Magnetic-field dependence of [(a) and (b)] the magnitudes of the lattice propagation vectors q_x^l and q_z^i, (c) integrated intensities of the incommensurate (closed circles) and commensurate (open circles) superlattice peaks, and (d) electric polarization along the *b*-axis (P_b) at 6 K. The magnetic field is applied along the *c*-axis. Reproduced from Ref. [4].

We have also investigated the effect of a magnetic-field on the superlattice reflection. When a magnetic field is applied along the c-axis at 6 K, $MnWO_4$ undergoes a ferroelectric transition at 3 T [see Fig. 3(d)]. Fig. 3 clearly demonstrates a sudden change in position as well as intensity of the superlattice reflection with increasing magnetic field. We conclude that the magnetic-field-induced ferroelectric transition at low temperature is accompanied by a magnetic transition from the commensurate collinear (AF1) to the incommensurate spiral (AF2) phase.

REFERENCES

[1] T. Arima, Y. Yamasaki, T. Goto, S. Iguchi, K. Ohgushi, S. Miyasaka and Y. Tokura, *J. Phys. Soc. Jpn.* **76**, (2007) 023602.
[2] K. Taniguchi, N. Abe, T. Takenobu, Y. Iwasa and T. Arima, *Phys. Rev. Lett.* **97**, (2006) 097203.
[3] G. Lautenschläger, H. Weitzel, T. Vogt, R. Hock, A. Böhm, M. Bonnet and H. Fuess, *Phys. Rev. B* **48**, (1993) 6087.
[4] K. Taniguchi, N. Abe, H. Sagayama, S. Ohtani, T. Takenobu, Y. Iwasa and T. Arima, *Phys. Rev. B* **77**, (2008) 064408.

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