**Direct observation of magnetic domain dynamics in Z-type cobalt hexaferrite**

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In the past decade, multiferroic materials with strongly coupled ferroelectric and ferromagnetic properties have received considerable attention, because it is considered to be promising for use in next-generation memory and novel spintronics devices [1,2]. However, it will be necessary to generate and control the magnetoelectric (ME) effects at room temperature and by low magnetic fields. Recently, large converse ME effects at room temperature was demonstrated in Z-type cobalt hexaferrite $(Ba,Sr)_3Co_2Fe_{24}O_{41}$ single crystal under low magnetic field [3]. Z-type cobalt hexaferrite ($Co_2Z$-hexaferrite) has a hexagonal lattice with centrosymmetric space group P63/mmc symmetry, composed of series of tetrahedral and octahedral Fe/Co layers of alternate stacks of L blocks and S blocks along c-axis. Therefore, strong low-field ME effects observed in $Co_2Z$-hexaferrite can be understood in terms of a transverse conical spin configuration of the magnetic building blocks at room-temperature. Although demonstration of mutual control of electric polarization (P) with magnetic field (H) and magnetization (M) with electric field (E) at room temperature in $Co_2Z$-hexaferrite was important breakthrough from both understanding and magnetoelectric devices application point of view [3], but the direct observation of magnetoelectric domain and their dynamics is crucial for further exposing their microscopic mechanism.

To achieve fundamental understanding of the nature of magnetoelectric state in $Co_2Z$-hexaferrite, we directly visualized magnetic domain dynamics in a $Co_2Z$-type hexaferrite as function of temperature and applied magnetic field, via in-situ transmission electron microscopy combined with the Lorentz transmission electron microscopy (LTEM). At room temperature, magnetic domains lie in *ab*-plane associated separated by Block-type domain walls. Magnetic domain is re-orientated from ferroxplane (*ab*-plane) to *c*-axis (~500K) as temperature increase (Fig.1), which is correlated with magnetic phase transition from transversal conical type to longitudinal conical spin type state. Furthermore, we observed biskyrmion spin texture under magnetic field (~0.1T) that were applied normal to sample plane (*ab*-plane) by control of objective lens current in TEM at the high-temperature region (~530K) (Fig.2). These results were consistent with the previous physical property results reported for this single crystal [3] and will present an important step toward the fundamental understanding of the nature of magnetoelectric state in $Co_2Z$-hexaferrite.

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Figure 1. (a) Lorentz TEM image in Fresnel mode at 300 K. (b-d) Magnetic induction map in $Co_2Z$-hexaferrite. Near the room temperature, easy magnetization axis is locked in *ab*-plane by in-plane anisotropy created from $Co^{2+}$. Magnetic easy axis rotates from the *ab*-plane to the *c*-axis as the temperature increase.

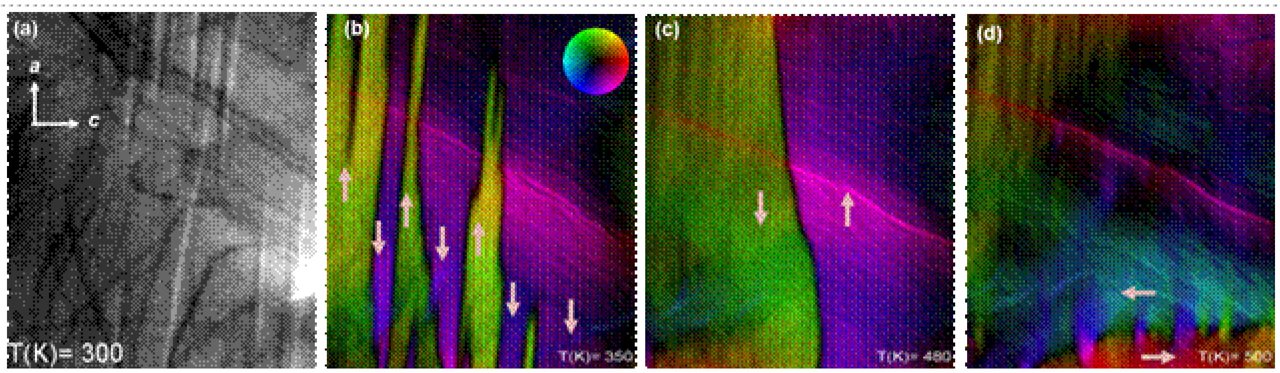

Figure 2. (a-c) Magnetic structure transition under magnetic field that were applied normal to sample plane at 530 K. (a) Helical stripe structure in the zero magnetic field. (c) Biskyrmion spin texture formed by applying magnetic field of 0.1 T. (d) Enlarged diagram of the biskyrmion crystal. Colors and arrows show the direction of electron spin in the biskyrmion.