Nano-scale local structural study of $BaTiO_3$ using STEM-CBED with a fast pixeletated STEM detector

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The convergent-beam electron diffraction (CBED) method has been applied for symmetry determination [1] and crystal structure and electrostatic potential analysis [2-4] of nano-scale local specimen areas. In order to examine specimens with local structural fluctuations, we proposed a combined use of scanning the transmission electron microscopy (STEM) and the CBED (STEM-CBED method [5]), which can be categorized into the 4D-STEM techniques. In the STEM-CBED method, CBED patterns are acquired pixel-by-pixel by scanning the convergent-beam electron probe with a sub-nanometer scan step, as schematically shown in Fig. 1(a). This method enables us to visualize nanometer-scale spatial distributions of local structures with picometer-scale sensitivity to atom-displacements with the full use of CBED data.

Using the STEM-CBED, we investigated local structures of perovskite-type ferroelectric oxides. Nano-scale fluctuations of the rhombohedral polarization clusters were visualized in the tetragonal and cubic phases of $BaTiO_3$ and $KNbO_3$ [5-8]. These results directly show the order-disorder-type characters in the structural phase transformations. However, long data acquisition time of STEM-CBED experiments restricted its wider applications.

In the present study, we have obtained STEM-CBED data of the tetragonal phase of $BaTiO_3$ using a transmission electron microscope JEM-ARM200F equipped with a fast pixelated STEM detector 4DCanvas [9]. The 4DCanvas detector with 264 x 264 pixels has a readout speed of 1000-4000 fps and a sensor quantum efficiency close to 100%, significantly reducing the acquisition time of STEM-CBED data. Figure 1(b) shows a CBED pattern acquired with an exposure time of 64 msec, showing a high s/n ratio enough for quantitative evaluation of symmetry breaking index $S = 100 \times (\sum_i \Delta I_i^2 / \sum_i I_i^2)^{1/2}$ [7-8]. Figure 1(c) shows a STEM-CBED map of the symmetry breaking index *S* of the tetragonal phase of $BaTiO_3$, where *S* was evaluated for the mirror symmetry along the *c**-direction which is expected in the tetragonal phase. Areas with higher values of S, where the tetragonal symmetry is broken, indicate the existence of the rhombohedral polarization clusters.

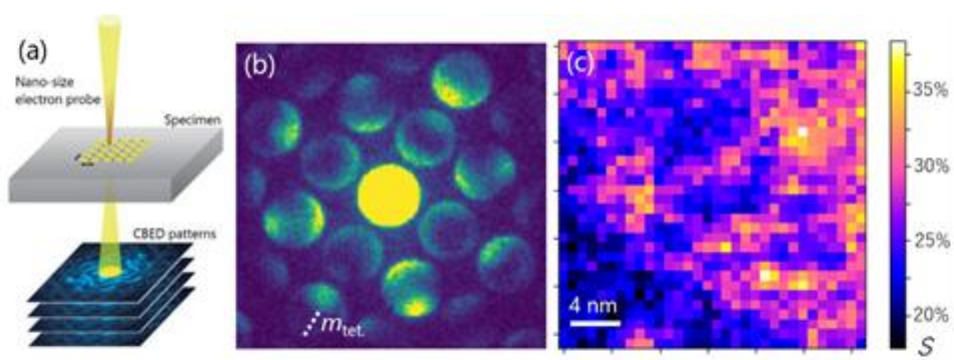

Fig. 1: (a) Schematic diagram of STEM-CBED. (b) CBED pattern of the ferroelectric tetragonal phase of $BaTiO_3$ acquired with the 4DCanvas detector. A white dotted line indicates the direction of the mirror symmetry expected in the tetragonal phase. (c) STEM-CBED map of symmetry breaking index *S*.

References

[1] e.g. M. Tanaka and K. Tsuda, J. Electron Microsc. **60** (Suppl. 1), S245 (2011).
[2] K. Tsuda and M. Tanaka, Acta Cryst. **A55** (1999), p. 939.
[3] Y. Ogata, K. Tsuda and M. Tanaka, Acta. Cryst. **A64**, 587 (2008).
[4] K. Tsuda, D. Morikawa, Y. Watanabe, S. Ohtani, T. Arima, Phys. Rev. B **81** (2010), p. 180102.
[5] K. Tsuda, A. Yasuhara and M. Tanaka, Appl. Phys. Lett. **102** (2013) p. 051913.
[6] K. Tsuda and M. Tanaka, Appl. Phys. Express **8**, (2015), p. 081501.
[7] K. Tsuda and M. Tanaka, Appl. Phys. Express **9**, 071501 (2016).
[8] K. Tsuda and M. Tanaka, Jpn. J. Appl. Phys. **56**, 10PB09 (2017).
[9] R. Sagawa et al., Microsc. Microanal. **23** (Suppl 1), 52 (2017).