

## Imaging 180° Polarization Reversal in Ferroelectric Oxides with Electron Backscatter Diffraction

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A ferroelectric material's permittivity is dependent on the extrinsic contribution of domain wall displacement during the application of an external field, which can involve the motion of both 90° and 180° domain walls. Directly quantifying non-180° domain structures through diffraction based techniques is generally straightforward; as the diffraction condition will be different from one side of a domain wall to the other, reflecting the rotation of the unit-cell and resulting in an easily quantifiable metric to determine location of domain wall [1]. Identifying 180° ferroelectric domains is more difficult, as Friedel's law states that the intensity of a diffracting plane and its inverse will be equal. Fortunately, Friedel's law is broken for dynamical scattering in a non-centrosymmetric crystal system, which is necessary for ferroelectricity. This has led to techniques such as convergent beam electron diffraction (CBED) being utilized for the characterization of 180° domain structures [1].

More recently, electron backscatter diffraction (EBSD) has been utilized to map 90° domain structures in ferroelectric oxides [2]. However, utilizing EBSD for imaging 180° domains has not been possible before, due to limitations in simulating dynamical EBSD patterns. Recently, however, De Graef has developed dynamical diffraction simulation software for EBSD, which we have used to map 180° polarization reversal in ferroelectric oxides [3].

In this work, we map 180° ferroelectric domains in periodically poled LiNbO<sub>3</sub> single crystals. Figure 1 demonstrates the difference in experimental patterns across a 180° domain boundary in LiNbO<sub>3</sub>. Figure 2 shows the simulated dynamical patterns of LiNbO<sub>3</sub> across a 180° domain boundary, with the same orientation as for the experimental patterns. Figure 3A is a secondary-electron SEM micrograph showing the 180° domain structures (contrast arises from a difference in the height of each domain variant). Along with the SEM image is traditional EBSD inverse pole figure (IPF) map, emphasizing that traditional indexing is not sensitive to polarization reversal. Figure 3C demonstrates the mapping of 180° domains using the dynamical diffraction simulations. Figure 3C, compares the simulated pattern from Figure 2a (positive polarization direction) to the patterns in the map from Figure 3A, by directly comparing the intensity differences between Figure 2a and the experimental patterns. The dark areas in Figure 3C correspond to the positive polarization direction. The opposite is true for Figure 3D, where the dark areas correspond to the negative polarization direction.

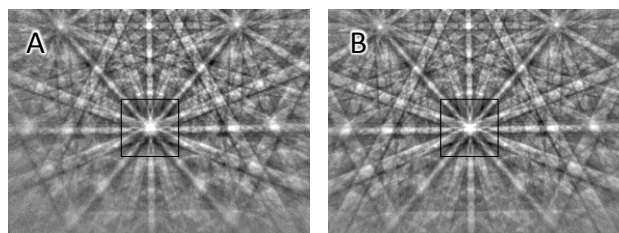
In conclusion, we demonstrate the ability to map 180° polarization reversal in ferroelectric oxides using EBSD. This new ability may lead to unprecedented characterization of domain structures in polar materials.

### References:

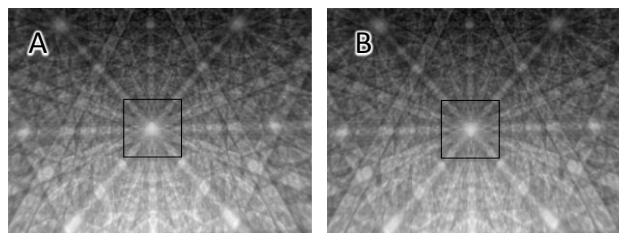
- [1] Y. H. HU, H. M. Chan, Z. X. Wen, and M. P. Harmer, *J. Am. Ceram. Soc.*, vol. 69, no. 8, pp. 594–602, 1986.
- [2] Howell, J. A., Vaudin, M. D., & Cook, R. F. (2014). *Journal of Materials Science*, 49(5), 2213-2224.

[3] P. G. Callahan and M. De Graef, *Microsc. Microanal.*, vol. 19, no. 05, pp. 1255–1265, 2013.

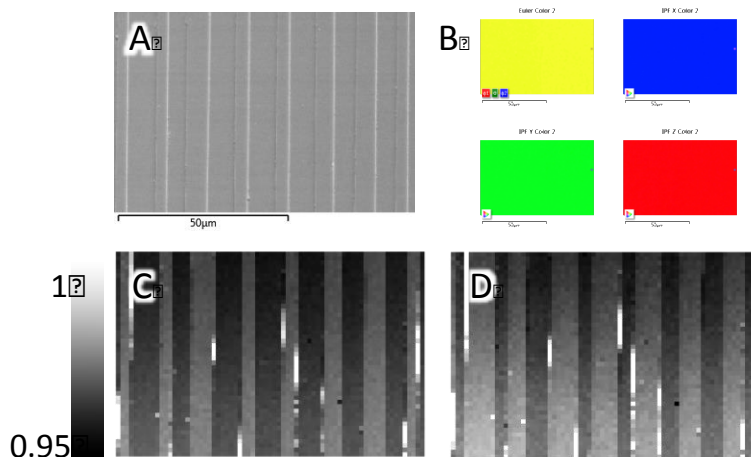
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**Figure 1.** Experimental EBSD patterns from  $\text{LiNbO}_3$  with (A) and (B) being inverse polarization directions and the inset boxes emphasizing the difference in the patterns.



**Figure 2.** Simulated EBSD patterns with the same orientations as Figure 1.



**Figure 3.** (A) An SEM micrograph of periodically poled  $\text{LiNbO}_3$  showing the  $180^\circ$  domain structures. (B) The corresponding IPF maps from EBSD, showing that traditional indexing is not sensitive to polarization inversion. (C) An intensity difference map comparing the experimental patterns from 3A to the simulated pattern shown in Figure 2A demonstrating  $180^\circ$  domains, with the dark regions having polarization directed out-of-the-page. (D) An intensity difference map comparing the experimental patterns from 3A to the simulated pattern shown in Figure 2B demonstrating  $180^\circ$  domains, with the dark areas having polarization direction into-the-page.