Determining Polarity in Ferroelectric Materials Using Precession Electron Diffraction and Cepstral Analysis

Colton Brown, Allison Mis Ph.D., Megan Holtz Ph.D.
Colorado School of Mines

Abstract
Understanding local crystallography is necessary for understanding ferroelectric behavior in materials. Key properties to know include lattice parameters, interatomic spacings, and polarity. One tool to learn these properties is scanning transmission electron microscopy (STEM), which can measure a diffraction pattern (kx, ky) at any scan position (x, y). This allows crystallographic properties to be investigated at the nanometer scale. However, diffraction signals can contain artifacts from sample thickness and misfit. These artifacts may be suppressed with precession electron diffraction (PED), which adds the additional benefit of increasing the number of reflections in the diffraction pattern. In this work, we perform analytical calculations to assess the efficacy of using PED combined with cepstral analysis to measure polarity in lead magnesium niobate-lead titanate (PMN-PT). The Fourier transform of the logistically scaled diffraction pattern yields the exit-wave power spectrum (EWPS) which yields interatomic spacings in the crystal. To extract polarity, we use the imaginary component of the transform which contains the antisymmetric information, which is called the exit-wave imaginary cepstrum (EWIC). The EWIC transform is improved by having higher-order diffraction disks, which is possible through PED. It was found that PED improves the EWIC signal: as precession angle was increased from 0° to 5°, signal noise decreased and dipole moments were better resolved in scenarios with and without misfit. These results indicate that cepstral analysis of real samples may benefit from PED.

Introduction
Ferroelectric materials are a type of polar material (the crystal structure has broken inversion symmetry [1] - see Fig. 1), and they have properties which make them viable for many electronic applications [2]. Understanding local crystallography is key to understanding their behavior, and Nano-beam electron diffraction (NBED) scanning transmission electron microscopy (STEM) is an effective way to measure local crystallographic parameters.

Cepstral Analysis

Electron diffraction patterns contain periodic signals that correspond to different crystallographic planes. The Fourier transform is a mathematical operation that converts periodic signals into distinct peaks. The exit-wave complex cepstrum (EWCC) is defined as:

\[ \text{EWCC} = FT(\text{FT}(V)) \]

Extracting the imaginary component of the EWCC yields the exit-wave imaginary cepstrum (EWIC), which contains only antisymmetric information. The broken symmetry of polar materials creates a dipole moment which can be quantified by the EWIC (Fig. 3).

Precession Electron Diffraction

Precession electron diffraction is a technique that rotates (precesses) an electron beam around a central axis to accumulate diffraction patterns under multiple diffraction conditions (Fig. 4). By rotating the electron beam, more higher-order diffraction disks are collected which improves the EWIC and EWPS transforms.

Methodology
Analytical calculations that simulated PED were performed using the Muller Group PC-STEM Package [3] for MATLAB to calculate diffraction patterns and their transforms.

To create sample misfit and precession effects, the Ewald’s sphere term was tilted. Sample misfit ranged from -10 mrad to 10 mrad on two axes of rotation (θx, θy) and precession angles up to 1° were tested. PED diffraction patterns at a given precession angle were created by using simulation diffraction at many tilts where the magnitude of perpendicular tilts θx, θy, and θz were equal to u.

Results
Increasing precession angle leads to clearer EWIC transforms. The dipole moment resolved, and noise is reduced. The improvement is seen in all tested sample misfit conditions (Figs. 5, 6, 7).

Conclusion
Analytical calculations were done to simulate electron diffraction with PMN-PT, to determine how using precession electron diffraction affected cepstral analysis. By using 1° of precession, noise in the EWIC is suppressed in conditions with and without sample misfit. Precession also significantly reduces angle and length measurement error that comes from sample misfit. The angle error is reduced by around a factor of 5, and the length error is reduced by around a factor of 2.5.