Effect of Zone Boundary Distortion on Improper Ferroelectrics

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In contrast to the proper ferroelectrics, in which the polarization is the order parameter, the electric energy is secondary in improper ferroelectrics. Therefore, in e.g., hexagonal $YMnO_3$, a prototypical improper ferroelectrics, absence of the critical thickness has been predicted: because the lattice distortion at the zone boundary is the order parameter, the accompanying polarization does not govern the overall stability.

$YMnO_3$ is unique in that the zone boundary distortion and the polarization can be separately monitored using the second-harmonic optical spectroscopy. Employing ultrathin films deposited on yttria-stabilized zirconia (111) substrates, we have unambiguously shown the evolution of the zone boundary distortion and the associated emergence of the polarization as the films grow thicker. This is the first experimental demonstration of the predicted lattice-polarization coupling in improper ferroelectrics.