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Uncovering In-Plane Domain Structures in Two-Dimensional Ferroelectric SnSe Using Machine-Learning Assisted 4D-STEM

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Ferroelectric two-dimensional (2D) materials have attracted growing attention because they retain a strong in-plane polarization down to the monolayer limit. Among these 2D ferroelectrics, group-IV monochalcogenides, including SnTe, SnSe, and SnS, contain strongly coupled in-plane ferroelectric polarization and spontaneous lattice strain [1]. In this work, we studied 2D SnSe using a machine-learning-assisted 4D-STEM approach developed in our group [2].

This approach utilizes an unsupervised learning algorithm to cluster diffraction patterns in 4D-STEM datasets. Based on the similarity of the diffraction patterns, the approach can distinguish the structural differences in the sample without prior knowledge. The algorithm uncovers super-domain structures in 2D SnSe (Figure 1b), where each super domain contains parallel stripes with different ferroelectric phases. By comparing the mean diffraction patterns of different domains, we identified their phase differences (Figure 1c) and lattice rotations (Figure 1d). These results further indicate the formation of twin boundaries between adjacent stripes, as well as a lattice rotation between super domains. The unsupervised learning analysis of the 4D-STEM data provides essential information on the atomic structure of each ferroelectric domain, paving the way for further quantitative study of the 2D SnSe.

To quantitatively map the domain structure, we calculated the reciprocal lattice parameters from the diffraction patterns using the center of mass (CoM) of each diffraction spot [3]. From the reciprocal lattice parameters, we extracted the in-plane ratio between the long and short edges of the orthorhombic lattice (Figure 2a). It indicates a twin domain boundary between two neighboring stripes, with their polarization along [100] and [010] directions correspondingly. In addition, we mapped the lattice rotation (Figure 2b) from the same 4D data, which suggests a ~2.5-degree difference between neighboring stripes, and a ~87.5-degree angle between two polarizations. This non-90-degree angle enables lattice-matched domain boundaries and avoids formations of dislocations or dangling bonds at the boundary. Meanwhile, we also observed a ~2-degree difference between the super domains, which leads to more complex lattice distortions within the areas where two super domains intersect (Figure 2c, d).

In conclusion, we utilized a machine-learning-assisted 4D-STEM approach to investigate 2D ferroelectric SnSe. The unsupervised learning uncovers the different phases of ferroelectric SnSe, while the subsequent quantitative analysis of the 4D data provides details of the lattice structure. Our method paves the way for a universal workflow for analyzing material deformations using 4D-STEM [4].

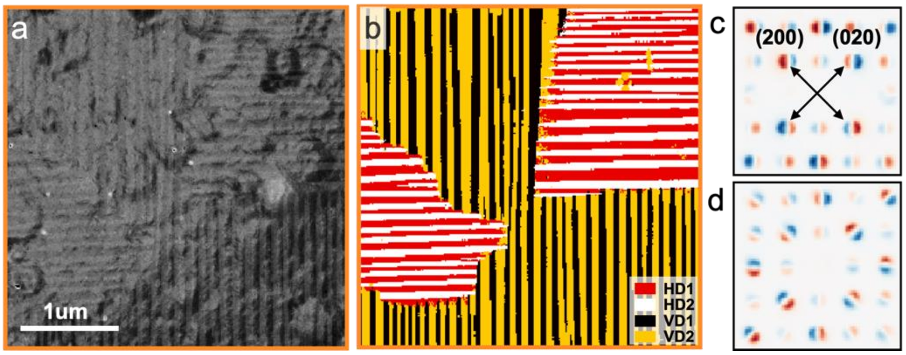

Figure 1 | Unsupervised learning of 4D data on 2D SnSe. (a) Virtual ADF image of a ferroelectric SnSe thin film. (b) Real-space map of four different clusters. (c) Difference between mean diffraction patterns from yellow and black stripes in (b), indicating vertical twin boundaries between these domains. (d) Difference between mean diffraction patterns from white and black stripes in (b), showing a lattice rotation between super domains.

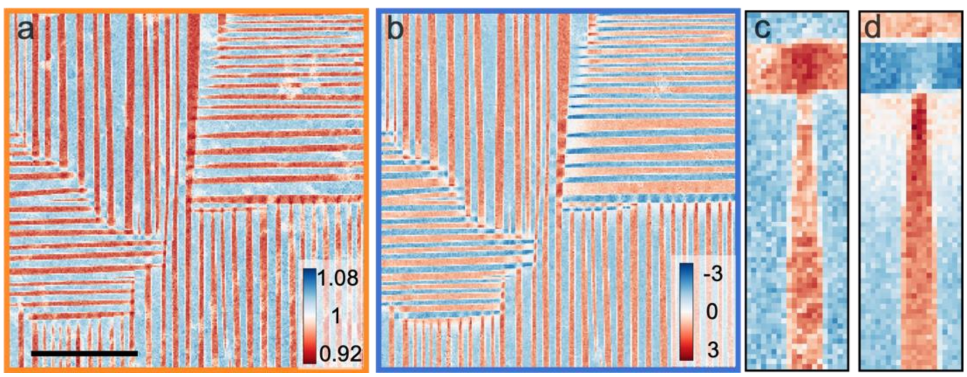

Figure 2 | Quantitative maps of phases and rotations from 4D data. (a) Phase map from the ratio between two in-plane lattice constants along [100] and [010]. (b) Lattice rotation map by measuring the rotation of [100] direction. (c-d) Phase map (c) and rotation map (d) at the boundary between two super domains.

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