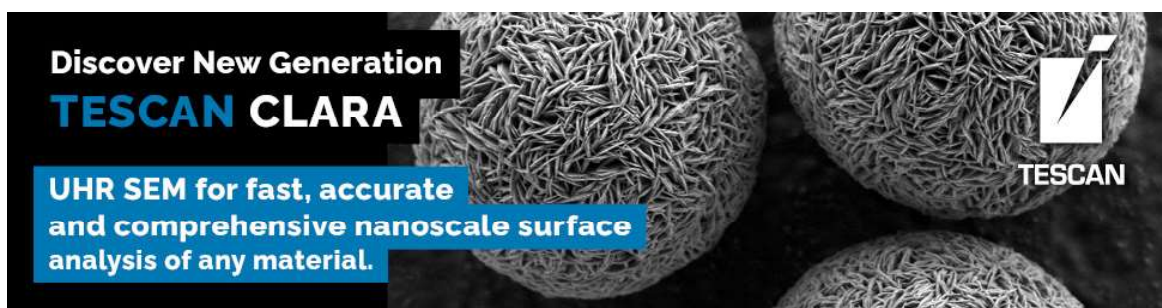


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Meeting-report

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In scanning transmission electron microscopy (STEM) imaging, information about local electrostatic potential – and by proxy, charge density – is inherently contained in the diffraction pattern at each scan position. Differential phase contrast (DPC) imaging has been used to image changes in electrostatic potential and local charge density [1], and center of mass (CoM) imaging has followed as a technique to further leverage the momentum information captured by pixelated detectors [2]. Local changes in charge density are particularly interesting in strongly charged domain walls (SCDWs), where screening charges accumulate and result in highly local increases in conductivity [3]. A nanoscale understanding of the precise distribution of charge across these domain walls is critical, yet it has remained difficult to extract the local charge distribution from other factors such as variations in sample tilt and sample damage.

In this work, we investigate SCDWs in out-of-plane polarized α -In₂Se₃ [4] using cross-sectional atomic resolution STEM, four-dimensional (4D) STEM, multislice simulations, and density functional theory (DFT). Recent years have seen the emergence of van der Waals ferroelectric materials, whose layered structure and stability down to few- and monolayer limits makes them particularly promising for nanoscale devices [5]. Here, we show that bending can be used to mechanically induce domain walls in α -In₂Se₃. This process can form both charge neutral transverse domain walls, which are oriented perpendicular to the basal plane of the material, and strongly charged lateral domain walls, oriented parallel to the basal plane (Fig. 1). Our atomic resolution imaging reveals charged domain walls with domain wall thicknesses of 1-2 nm. With 4D-STEM and center of mass imaging, we measure the charge density across these charged interfaces. We observe layers of bound and free charge at the domain wall, which are sharply localized to within 3-4 nm of the interface (Fig. 2). With DFT, we also present a theoretical basis for the electronic structure arising at these interfaces. Using the hybrid HSE06 functional, our DFT simulations predict the emergence of a mid-gap electronic state, localized to the domain wall, where charge carriers are free to accumulate. The DFT-predicted charge states complement our simulated 4D-STEM data, which we conducted using the multislice method [6]. These methods combined show the highly localized SCDWs in α -In₂Se₃ and demonstrate a potential methodology for studying local electronic properties at ferroelectric interfaces in van der Waals materials [8].

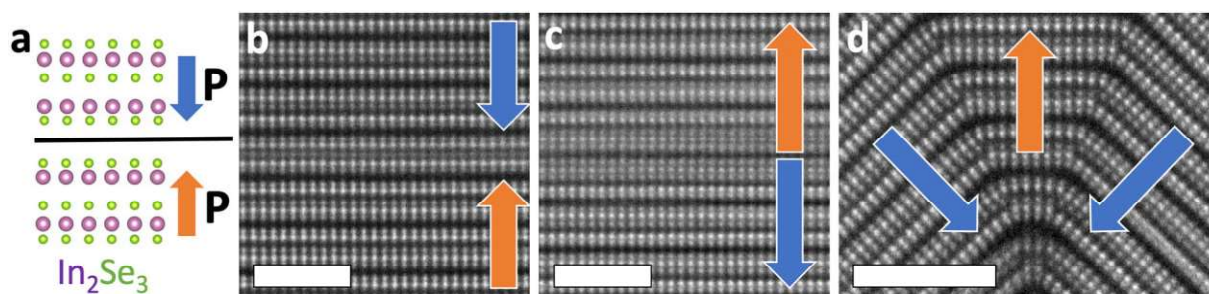


Fig. 1. Atomic structure of bend-induced domain walls in α -In₂Se₃. a) Cartoon model of oppositely polarized α -In₂Se₃ monolayers. b-d) HAADF-STEM images of b) a head-to-head lateral domain wall, c) a tail-to-tail lateral domain wall, and d) two adjacent transverse domain walls. All scale bars are 2 nm.

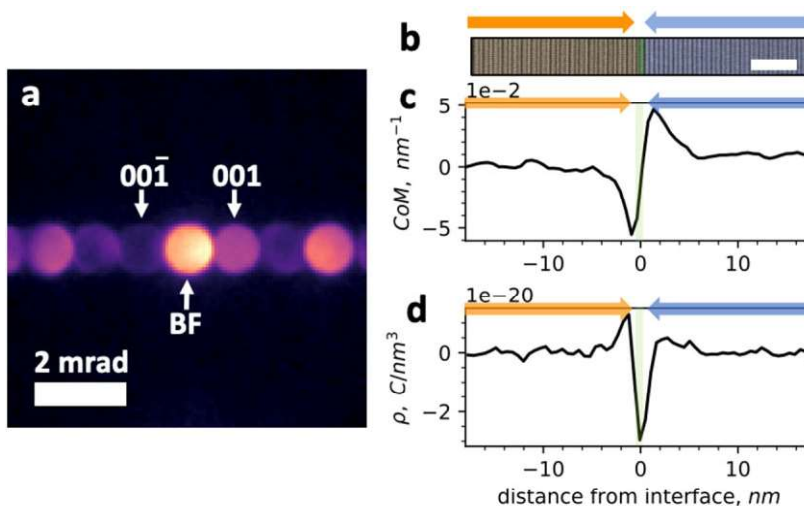


Fig. 2. Charge density measurement across a bend-induced head-to-head strongly charged domain wall. a) Representative CBED for region immediately to the right of the domain wall from 4D-STEM data acquired using EMPAD [7]. b) HAADF-STEM image of a head-to-head charged domain wall, correlating to scan region for c & d. Scale bar 5 nm. c) Averaged CoM deflection of the bright field disk across a head-to-head domain wall with polarization direction indicated by arrows. d) Charge density across the domain wall, as measured from the CoM deflections in (c).

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