Electron Ptychography Resolves Individual Atomic Layers in Twisted Super-Moiré

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Twisted moiré systems have attracted considerable attention due to their tunable electronic and optical properties enabled by interlayer coupling modulated by the twist angle, leading to emergent phenomena such as superconductivity, correlated insulating states, and unconventional ferroelectricity [1]. While most studies have focused on small-angle twisted systems, a new supermoiré lattice has been identified near large commensurate twist angles, exhibiting robust periodicity and flat-band engineerability [2]. Unlike conventional moiré lattices formed by simple AA or AB stacking patterns at small twist angles, the supermoiré domains arise from more complex commensurate stacking arrangements, referred to as commensurate rotational faults (CRFs) [3], making their characterization particularly challenging. In this work, we employ multislice electron ptychography [4] to clearly resolve the individual layer structures of bilayer–bilayer twisted WSe_2 near the 21.8° commensurate angle, successfully identifying three distinct CRF structures that collectively form the supermoiré lattice.

The schematic of electron ptychography is illustrated in **Fig. 1a**, where a defocused electron probe scans across the bilayer–bilayer WSe_2 sample, with each bilayer having an approximate thickness of 1.5 nm. The resulting diffraction patterns are recorded using an electron microscope pixel array detector (EMPAD). In the reconstruction process, multiple mixed-state probe modes [5] are employed to accurately account for the partial coherence of the electron probe, thereby enhancing resolution. Unlike conventional multislice ptychography reconstruction, regularization is omitted to fully distinguish the two independent bilayers with different orientations. As a result, the reconstructed slices clearly separate the top and bottom bilayers, with each slice containing only a single hexagonal lattice of uniform orientation (**Fig. 1b-c**). By overlapping the two reconstructed slices, the twisted lattice structure is distinctly visualized (**Fig. 1d**), providing both high lateral resolution and layer-resolved information.

To clearly visualize the supermoiré structure, the inverse fast Fourier transform (iFFT) of the first-order FFT spots (inset **Fig. 2a**), which correspond to the 21.8° moiré frequency, is generated and overlaid with the twisted lattice (**Fig. 2a**). A representative supermoiré domain, formed by three commensurate rotational faults (CRFs), is labeled with four sublattice exchange odd site centering around W atoms (SE_{odd_W}) located at the corners, and SE_{even} and SE_{odd_Se} sites appear in the middle of the domain. Zoomed-in images of the three CRFs, along with their corresponding lattice schematics, are presented in **Fig. 2b–d**, providing a detailed view of the atomic structures. Although the relatively large 21.8° twist angle results in a small moiré wavelength of only 0.86 nm, the supermoiré structure exhibits a significantly larger periodicity of 7.2 nm. This scale is comparable to the wavelength of small twisted angle systems, highlighting the potential of large twisted angle supermoiré lattices for exploring the interplay among band topology, quantum geometry, and moiré superconductivity [6].

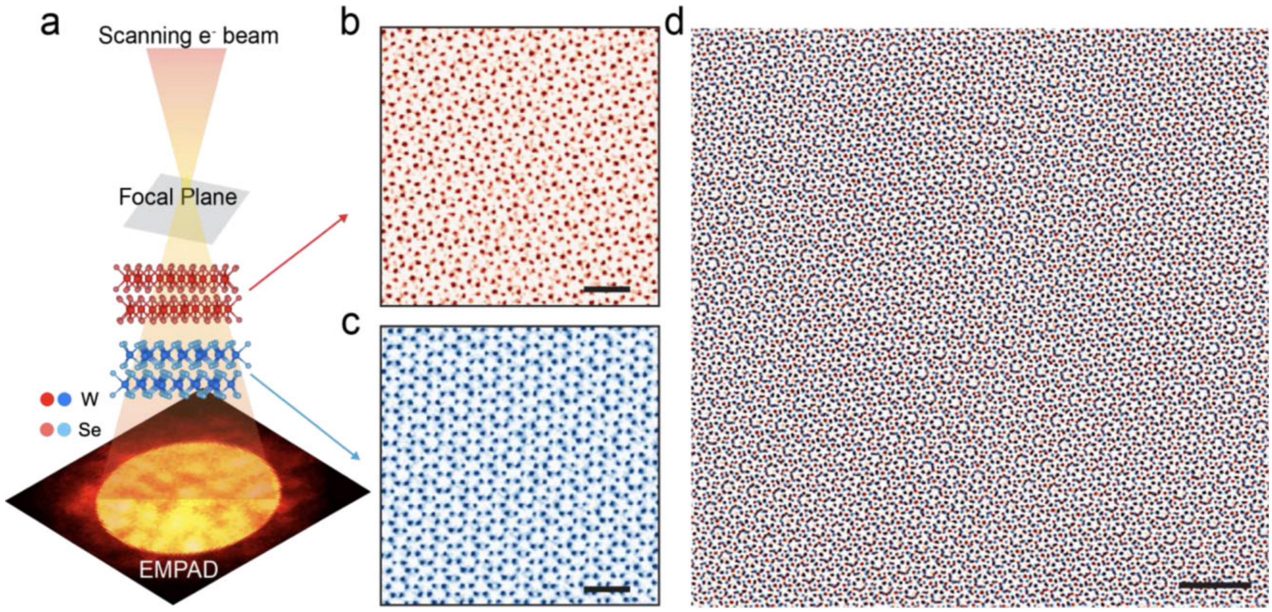

Fig. 1. (a) Schematic of the multislice electron ptychography on bilayer-bilayer twisted WSe_2. (b-c) Decoupled top (b) and bottom (c) layers. Scale bar: 1nm. (d) Twisted lattice image overlapped by the top (red) and bottom (blue) layer. Scale bar: 2nm.

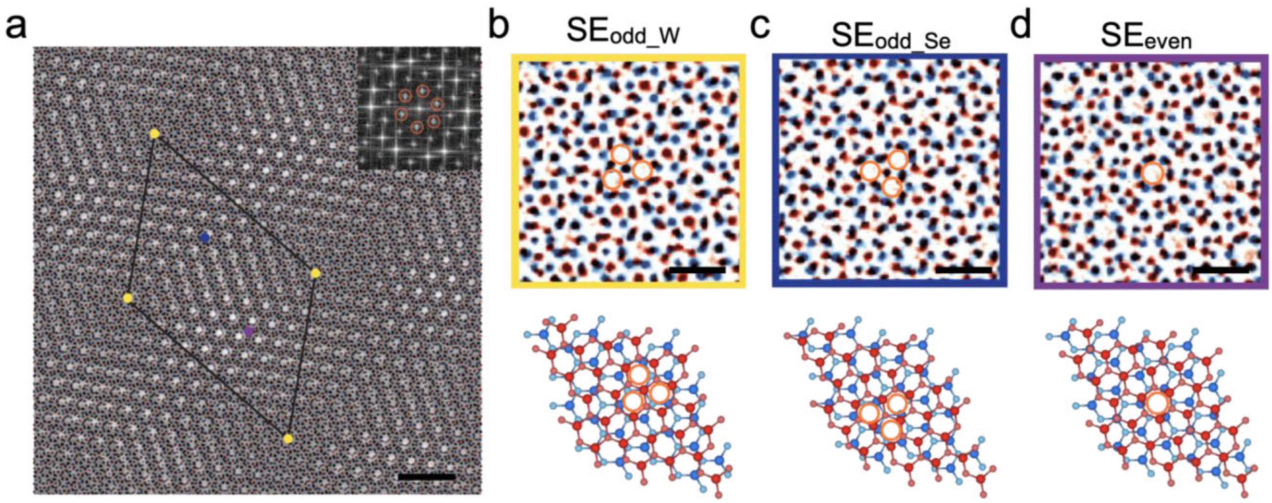

Fig. 2. (a) Twisted lattice overlapped with the iFFT image. The super moiré domain is labeled with four SE_{odd_W} (yellow) at the corner and SE_{odd_Se} (blue) and SE_{even} (purple) in the middle. FFT image is inset with the first order spots labeled in orange circles. Scale bar: 2nm. (b-d) Zoomed in images and lattice schematic of the three CRFs. Scale bar: 0.5nm.

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