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Strain Relaxation and Excitonic Absorption of Atomically-Reconstructed WSe_2 Moiré Superlattices

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Atomically-thin layers of semiconducting transition metal dichalcogenides (TMDs), namely tungsten diselenide (WSe_2), have attracted considerable interest because of new properties that can be obtained when artificially fabricated into van der Waals superlattices. Interlayer coupling is responsible for various properties, including the indirect-to-direct band gap cross-over from bulk and few-layered to monolayer TMDs. It has also been demonstrated that twist angle can be used to tune the interlayer coupling strength in bilayer MoS_2 [1] and WS_2 [2] homostructures. In this work, the high spatial and spectral resolution of monochromated electron energy-loss spectroscopy (EELS) coupled with four-dimensional scanning transmission electron microscopy (4D-STEM) in an aberration-corrected STEM are used to investigate the excitonic response of twisted bilayer WSe_2 with a low moiré angle and the strain relaxation of those that are atomically reconstructed.

Monolayered WSe_2 flakes were mechanically exfoliated from synthetic bulk crystals and transferred onto SiO_2/Si wafers prior to assembly. The twisted bilayer WSe_2 structures were fabricated using a viscoelastic 'tear and stack' stamping method for controlled stacking and twist angle [3], before depositing onto a Quantifoil holey carbon TEM grid. Monochromated EELS and nano-beam electron diffraction with 1 mrad convergence angle were performed on a modified Nion HERMES-S200 (or ChromaTEM) operated at 60 kV with the sample cooled using liquid nitrogen (T ~ 150 K).

Well-defined hexagonal moiré patterns with nanometer periodicity are apparent for the low twist angles with high-resolution imaging. At twist angles below ~3°, some of the high-symmetry stacking regions become more energetically favorable, leading to an expansion of selected high-symmetry points into domains by atomic reconstruction. A low-angle annular dark-field (LAADF) image can be used to visualize the domain boundaries [4]. The domain boundary geometry has been demonstrated to differ between those of R-type (AA) and H-type (AA') stacking order in twisted TMDs that designates layers without and with inversion symmetry, respectively. R-type takes on a triangular geometry [Fig. 1(b,c)], while H-type resembles a kagome-like pattern [Fig. 1(d)]. 4D-STEM of the reconstructed bilayers reveals the nanoscale deformation that correlates perfectly with the location of the domain boundaries [Fig. 2]. The excitonic absorption signatures of these nanometric twisted bilayers [Fig. 1(e)] demonstrate a good general correspondence to the thickness dependence of the exciton resonances, namely a pronounced decrease in C exciton energy with the number of layers [5]. Comparing different twist angles in the bilayers also shows sizable blueshifts in the C exciton energy up to 200 meV, with extremes between the zero-twist and towards the anti-aligned (28°) case, indicating underlying differences in interlayer coupling with respect to the moiré angle [6].

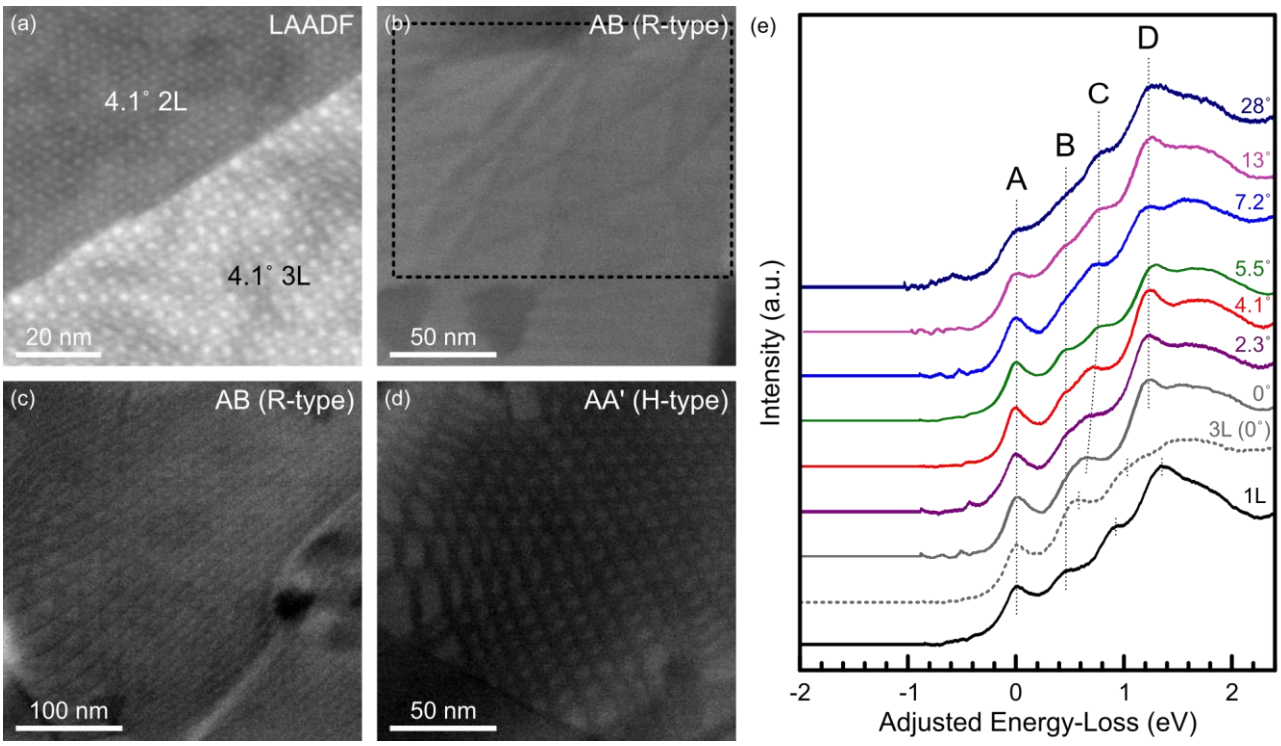

Figure 1. STEM-LAADF images of WSe_2 twisted bilayers (2Ls) with (a) 4.1° twist angle, and (b,c) AB (R-type) stacking, (d) AA' (H-type) stacking with atomic reconstruction. (e) Monochromated EELS spectra from twisted bilayers with various moiré angles compared to a representative monolayer (1L) and trilayer (3L) with zero twist. Spectra are offset in energy relative to the A exciton, and the dotted lines are to help guide the eye in the different exciton energy positions.

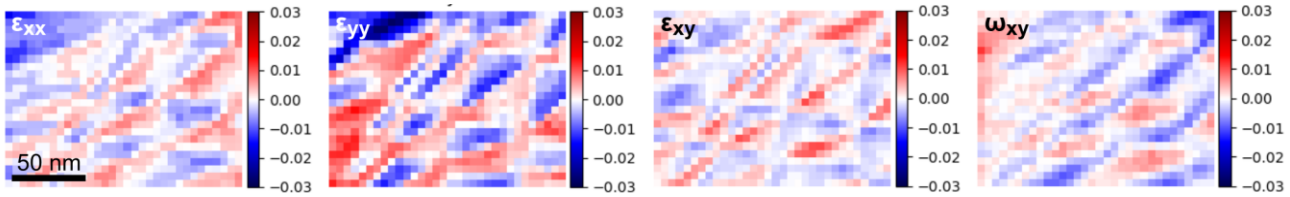

Figure 2. Strain maps obtained using 4D-STEM of the boxed region from Fig. 1(b), including axial strains, ε_{xx}, ε_{yy}, shear strain, $\varepsilon_{xy,}$ and rotation $\omega_{xy,}$ showing strain relaxation corresponding to the distribution of alternating AB/BA domains as a result of the atomic reconstruction.

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