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Twist and Bend in Van Der Waals Materials and 2D Stacked Heterostructures

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The rapidly growing field of van der Waals materials and 2D heterostructures continues to offer surprises to researchers. Here we will describe the use of high resolution annular dark field scanning transmission electron microscopy (STEM) to uncover new structural features in this exciting class of material. For example, we will present a systematic cross sectional STEM investigation of the atomically flat interfaces between transition metal dichalcogenides and boron nitride. These are crucial in many optoelectronic device architectures but we find that unless the selenides (MoSe2, WSe2, NbSe2) are processed in an inert environment protruding defects prevent the realisation of pristine interfaces.[1] Cross sectional imaging can also be harnessed to reveal the rich microstructures that may be produced when layered van der Waals materials are subjected to mechanical deformation. Using graphite, BN and MoSe2 as examples, we show that the types of structural defects produced can be predicted based on knowledge of the bend angle and thickness of the material.[2] We find that above a critical thickness the materials exhibit numerous twin boundaries and that for large bend angles these can contain nanoscale regions of local delamination. We show that the induced changes in local stacking order could be used to locally tune the optoelectronic behaviour and propose that such bend features are important in determining how easily the material can be thinned in the early stages of mechanical or liquid exfoliation.

In addition, we will discuss new results revealing the structural relaxation that occurs when two transition metal dichalcogenide (TMDC) monolayers are misoriented by a small twist (rotation angle). Others work has shown that graphene bilayers reconstruct at small twist angles giving local regions of perfect stacking bounded by partial dislocations [3]. However, many high profile experimental measurements of the exciting physics present in twisted TMDCs in 2019 has ignored the potential for local structural relaxation. Here we demonstrate using atomic resolution STEM that TMDCs bilayers twisted to a small angle, less than ~3˚, reconstruct into energetically favourable stacking domains separated by a network of stacking faults, and that this behaviour is more complex and more powerful than is observed in graphene sheets [4]. At low magnification these structures are visible by optimising dark field imaging condition (Figure 1) but we also demonstrate their analysis at the atomic scale using patch based PCA denoising to improve the signal to noise ratio (Figure 2). For crystal alignments close to 3R stacking, we find that a tessellated pattern of mirror reflected triangular 3R domains emerges, separated by a network of partial dislocations which persist to the smallest twist angles. Complementary scanning tunneling measurements show that the electronic properties of these 3R domains appear qualitatively different from 2H stacked TMDs, featuring layer-polarized conduction band states caused by lack of both inversion and mirror symmetry. In contrast, for alignments close to 2H stacking, stable 2H domains dominate, with nuclei of an earlier unnoticed metastable phase limited to ~ 5nm in size. This appears as a kagome-like pattern at twist angles less than 1 ˚, transitioning to a hexagonal array of screw dislocations separating large-area 2H domains as the twist angle approaches 0 ˚. Tunneling spectroscopy measurements reveals that such reconstruction creates strong piezoelectric textures and pseudo-magnetic fields, opening new avenues for engineering of 2D material properties on the nanometre scale.

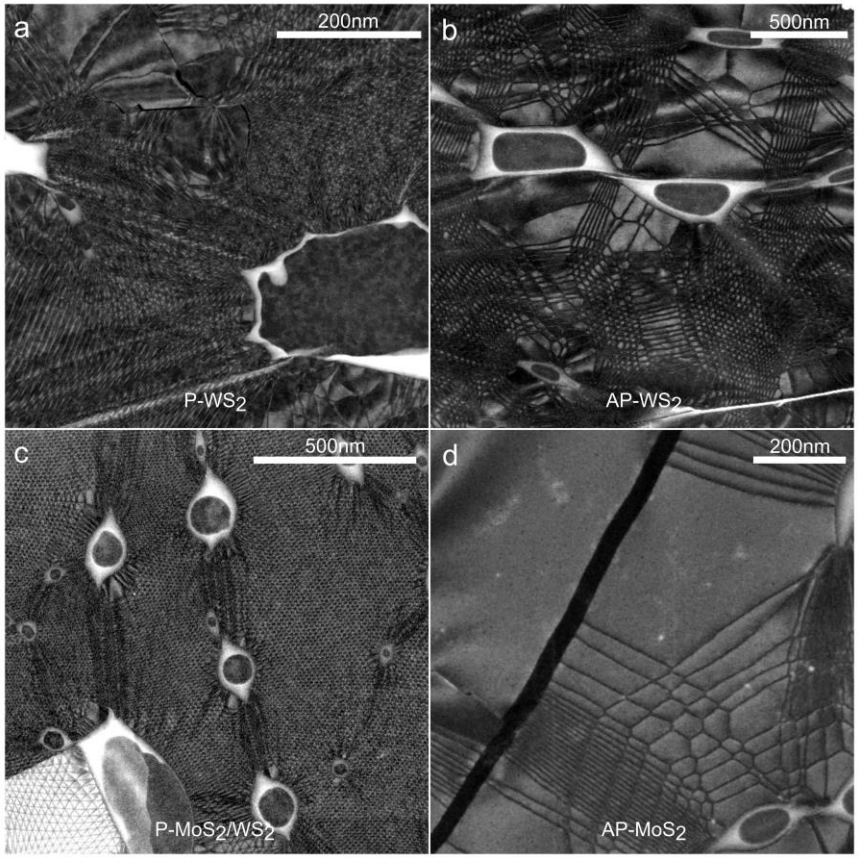

Figure 1. Low magnification ADF STEM images comparing lattice domain formation in homo- and hetero-bilayers of MoS2 and WS2 with parallel stacking alignments (close to 3R) or anti-parallel alignment (close to 2H). Reproduced from reference [4].

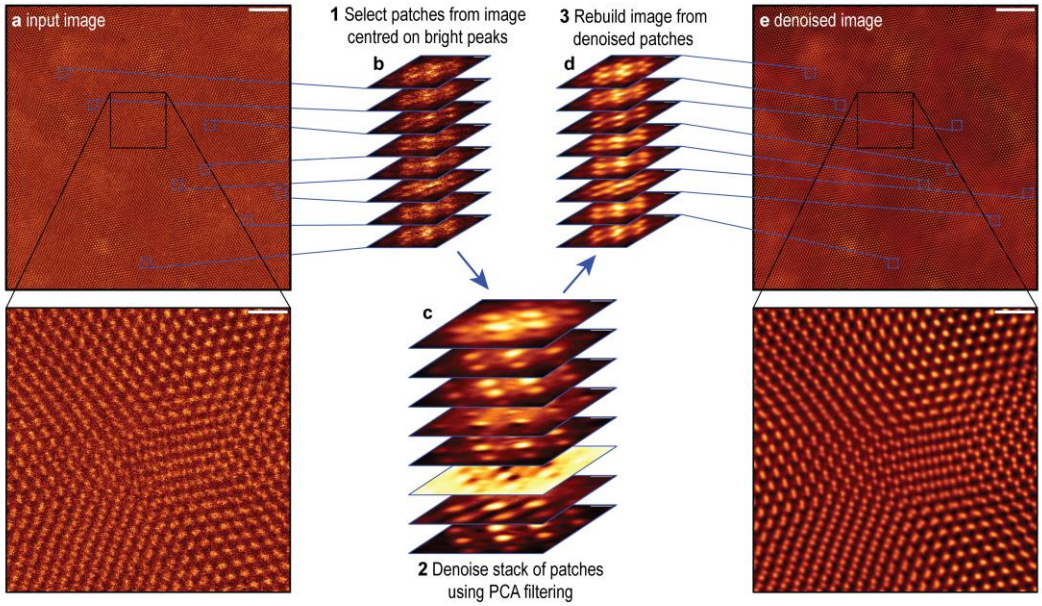

Figure 2. Illustration of patch based principle component analysis (PCA) filtering process using the FastICA algorithm. a) shows the raw image which was processed into figure 2d in the main text. The lower panel shows a magnified region of the upper panel. b 8 example image patches (65 x 65 px) extracted

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from the locations indicated by the blue squares in a. In this case 18831 patches were used. c) First 8 PCA loadings (of 4000+) obtained using FastICA algorithm on stack of all patches from b. d) 8 example patches after reconstruction using 16 PCA components. e) Reconstructed image built from PCA filtered patches. Scale bars (a,e) top 5 nm, bottom 1 nm, (b-d) are square regions 1.25 nm x 1.25 nm. Reproduced from reference [4].

References

[1] A.P. Rooney et al. Observing Imperfection in Atomic Interfaces for van der Waals Heterostructures, Nano letters 17, 5222, (2017)
[2] A.P. Rooney et al. Anomalous twin boundaries in two dimensional materials, Nature communications 9, 3597, (2018)
[3] Yoo, H. et al. Atomic and electronic reconstruction at the van der Waals interface in twisted bilayer graphene. Nature Materials 18, 448–453 (2019).
[4] A. Weston et al, Atomic reconstruction in twisted bilayers of transition metal dichalcogenides, Nature Nanotechnology, in press (2020)

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