“Low voltage aberration corrected STEM for 2D materials”
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Aberration-corrected scanning transmission electron microscopy (STEM) operated at low accelerating voltage can now provide real space imaging and spectroscopy analysis at the atomic scale with single atom sensitivity. This opens new opportunities for quantitative study of the structure and chemistry in 2D materials and monitoring of their dynamical behavior under e-beam irradiation or in-situ heating. Such studies, especially when combined with first-principles calculations, serve as an important step to correlate the atomic structure with their local properties, unveil the atomic growth mechanism for new quantum-structures, and help to create new functionalities in these 2D materials via defect engineering.

Using low-voltage aberration corrected STEM imaging, we show that for epitaxial grown 2D lateral heterostructures where the two monolayer components have similar crystal structure but different lattice constants, such as WS$_2$/WSe$_2$ or MoS$_2$/MoSe$_2$, strain relaxation at lateral interfaces due to lattice mismatch often lead to misfit dislocation arrays. We demonstrate that such misfit dislocations can induce the formation and growth of sub-2-nm quantum-well arrays in semiconductor monolayers, driven by dislocation climb. This misfit-dislocation-driven growth can be applied to different combinations of 2D monolayers with lattice mismatch, paving the way to a wide range of 2D quantum-well superlattices with controllable band alignment and nanoscale width.

By performing in-situ annealing experiments in STEM, we found that stacking faults and rotational disorders in multilayered 2D crystals can be healed by grain boundary (GB) sliding, which works like a “wiper blade” to correct all metastable phases into thermodynamically stable phase along its trace. The driving force for GB sliding is the gain in interlayer binding energy as the more stable phase grows at the expanse of the metastable ones. Density functional theory calculations show that the correction of 2D stacking faults is triggered by the ejection of Mo atoms in mirror twin boundaries, followed by the collective migrations of 1D GB. The study highlights the role of the often-neglected interlayer interactions for defect repair in 2D materials and shows that exploiting these interactions has significant potential for obtaining large-scale defect-free 2D films.

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