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Spatially Correlated Disorder in Epitaxial van der Waals Heterostructures¹ NOUAMANE LAANAIT, Center for Nanophase Materials Sciences, Oak Ridge National Lab, ZHAN ZHANG, Argonne National Lab, CHRIS-TIAN SCHLEPUTZ, Paul Sherrer Institute, YING LIU, University of Wisconsin, Milwaukee, MICHAEL WOJCIK, Argonne National Lab, RACHAEL MYERS-WARD, D. KURT GASKILL, U.S Naval Research Lab, PAUL FENTER, Argonne National Lab, LIAN LI, University of Wisconsin, Milwaukee — The structural cohesion of van der Waals (vdW) heterostructures relies upon a cooperative balance between strong intra-layer bonded interactions and weak inter-layer coupling. The confinement of extended defects to within a single vdW layer and competing interactions introduced by epitaxial constraints could generate fundamentally new structural disorders. Here we report on the presence of spatially correlated and localized disorder states that coexist with the near perfect crystallographic order along the growth direction of epitaxial vdW heterostructure of Bi2Se3/graphene/SiC grown by molecular beam epitaxy. With the depth penetration of hard X-ray diffraction microscopy and high-resolution surface scattering, we imaged local structural configurations from the atomic to mesoscopic length scales, and found that these disorder states result as a confluence of atomic scale modulations in the strength of vdW layer-layer interactions and nanoscale boundary conditions imposed by the substrate. These findings reveal a vast landscape of novel disorder states that can be manifested in epitaxial vdW heterostructures.

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