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Van der Waals Epitaxy of Ultrathin Halide Perovkistes YIPING WANG, YUNFENG SHI, JIAN SHI, Department of Material Science and Engineering, Rensselaer Polytechnic Institute — We present our understanding, with $CH_3NH_3PbX_3$ as a model system, on the 2D van der Waals growth and kinetics of 3D parent materials. We show the successful synthesis of ultrathin (sub-10 nm), large scale (a few tens of μm) single crystalline 2D perovskite thin films on layered mica substrate by van der Waals (VDW) epitaxy. Classical nucleation and growth model explaining conventional epitaxy has been modified to interpret the unique 2D results under VDW mechanism. The generalization of our model shows that a 3D crystal with low cohesive energy tends to favor the 2D growth while the one with strong cohesive energy has less kinetic window. With Monte Carlo simulations, we show that the fractal 2D morphology in perovskite precisely manifests the kinetic competition between VDW diffusivity and thermodynamic driving force, a unique phenomenon to VDW growth, suggesting a fundamental limit on the morphology stability of the 2D form of a 3D material. On the other hand, our single crystal thin film growth results and subsequent cryogenic study in the iodide perovskite provide a perfect resource for the exploration of its complex optical and electronic properties and unveiling the origins of its popularity in the energy conversion field.

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