

# Novel Growth Mechanisms in van der Waals Epitaxy: 3D Morphologies of $\text{Bi}_2\text{Se}_3$

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Van der Waals (vdW) materials, such as graphene, transition metal dichalcogenides, and bismuth chalcogenides, are an increasingly popular research topic due to their wide variety of applications. They are characterized by strongly bonded layers in the  $a$ - $b$  plane and weak vdW bonds between layers in the  $c$ -direction. Careful control of vdW materials could create designer stacks of 2D materials or unlock the unique physics of 3D topological insulators, resulting in improved optoelectronic, spintronic, and valleytronic technologies. Growth via Molecular Beam Epitaxy (MBE) is a promising avenue for improved material quality due to its' highly customizable growth conditions. Unlike traditional MBE, which relies on the strong interaction between the growth material and a lattice matched substrate, vdW epitaxy takes advantage of the weak vdW bonds to grow on substrates with vastly different crystal structures. Unfortunately, vdW epitaxy cannot be fully explained via the well-known mechanics of traditional MBE. Growth of atomically smooth layers remains challenging as vdW materials tend to grow in terraced “wedding cake” morphologies. Also, vdW materials favor the (001) orientation with the vdW gaps parallel to the growth surface. Other orientations have been reported via MBE but require extensive substrate pretreatment or pre patterning. Improved understanding of the growth mechanisms involved in vdW epitaxy could allow for improved material quality as well as exotic growth morphologies.

In this study, we use the topological insulator  $\text{Bi}_2\text{Se}_3$  as a prototypical vdW material for the exploration of growth mechanics. We chose to grow on GaAs(001) substrates as the interaction of adatoms with the post deoxidization reconstructed surface may provide greater insight than a less complex growth surface, such as previous work on sapphire substrates. It was found that growth at a substrate temperature of 425°C produced a significant amount of growth in the elusive (0015) orientation, with domains aligned along the GaAs[110] axis (Sample A). It is believed that bismuth aggregation due to thermal degradation as well as fast diffusion of bismuth along GaAs[110] combine to push growth into this orientation. A film grown on a 10nm  $(\text{Bi}_{0.5}\text{In}_{0.5})_2\text{Se}_3$  buffer layer results in the formation of  $\text{Bi}_2\text{Se}_3$  platelets with

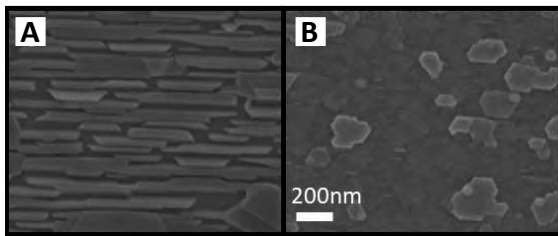


Figure 1: SEM images of  $\text{Bi}_2\text{Se}_3$  films grown on GaAs substrates with varying growth parameters.

sheer walls and flat tops (Sample B). Based on this research it is believed that vdW growth dynamics are dominated by the relative strength of film/adatom and substrate/adatom interactions. The morphologies reported here differ significantly from those reported on sapphire, implying that substrate interaction is key to understanding novel morphologies in vdW materials.

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## Supplementary Pages (Optional)

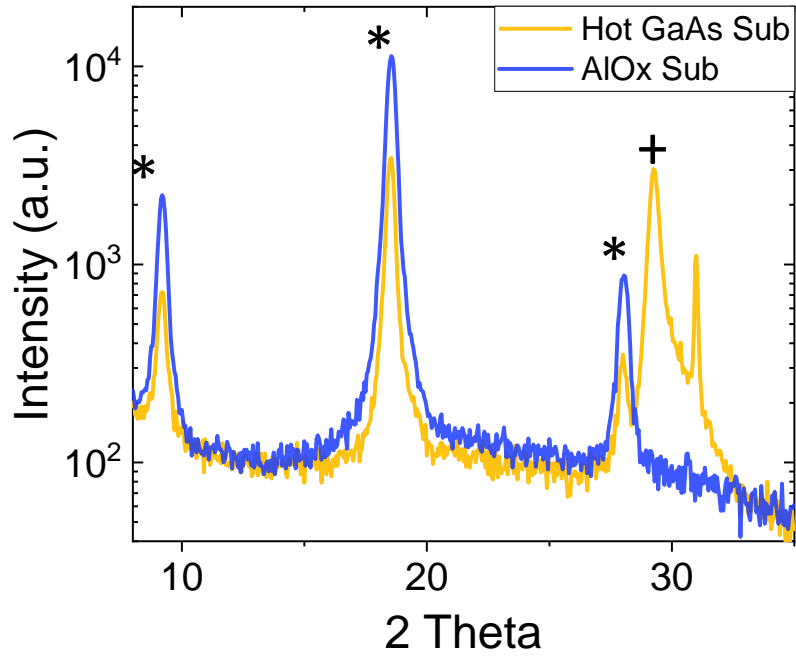


Figure S1: XRD scans confirming the presence of the (0015) orientation of  $\text{Bi}_2\text{Se}_3$  grown on a GaAs(001) substrate at high temperature. The  $\text{Bi}_2\text{Se}_3$  (0001) peaks are indicated with a \*, while the (0015) peak is indicated with +.