

Formation of twin-free single phase β - In_2Se_3 layers via selenium diffusion into $\text{InP}(111)\text{B}$ substrate

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Indium selenide, In_2Se_3 , has recently attracted growing interest due to its remarkable properties, including room temperature ferroelectricity, outstanding photoresponsivity, and exotic in-plane ferroelectricity, which open up new regimes for next generation electronics [1]. Yet, In_2Se_3 has many different polymorphs [2], and it has been challenging to synthesize single-phase material, especially using scalable growth methods, as needed for technological applications. We recently reported the growth of twin-free ultra-thin layers of In_2Se_3 prepared by a diffusion driven molecular beam epitaxy approach, and twin-free Bi_2Se_3 layers grown on these unique virtual substrates [3].

In this study, we use aberration-corrected scanning transmission electron microscopy (STEM) to characterize the microstructure of these materials and understand the growth mechanism of twin-free In_2Se_3 layers. We show

that high quality In_2Se_3 and Bi_2Se_3 crystalline layers that are fully twin-free and largely free of defects, can be achieved using $\text{InP}(111)\text{B}$ substrates. STEM observations also show that the sample consists primarily of single phase β - In_2Se_3 (Figure 1a). Further, the contrast difference visible in cross-sectional bright field (BF) STEM image (Figures 1b), just below the $\text{In}_2\text{Se}_3/\text{InP}$ interface suggests significant Se diffusion into the substrate beyond the $\text{In}_2\text{Se}_3/\text{InP}$ interface. However, the excess Se does not alter the crystal structure of the InP substrate, which remains zinc blende. This observation suggests that the In atoms remain fixed in their lattice sites while Se diffuses into the substrate, displacing P. Then the lattice transforms into the stable rhombohedral In_2Se_3 structure. This result implies that In atoms are not mobile during the transformation, thus resulting in twin-free In_2Se_3 . This mechanism for the In_2Se_3 formation also explains the resulting pure β -phase In_2Se_3 and may enable the possibility of stabilizing other technologically interesting phases of In_2Se_3 .

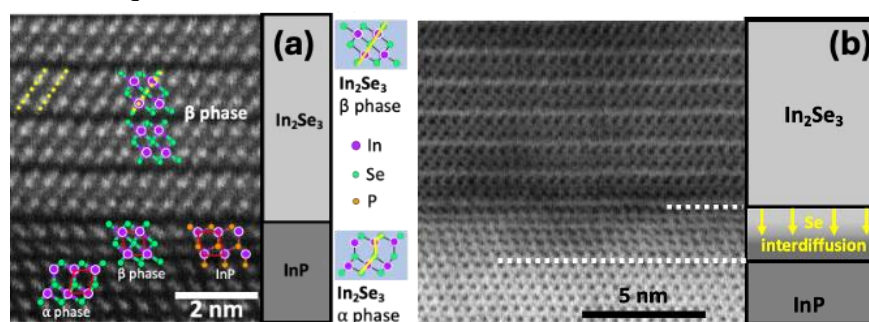


Figure 1. (a) atomic resolution of the β phase In_2Se_3 layer. (b). Cross-sectional BF-STEM image at higher magnification showing that the presence of excess Se atoms does not alter the crystal layers.

[1] P. Ajayan, P. Kim, K. Banerjee, Phys. Today. **69**, 38-44(2016)

[2] N. Balakrishnan, E. D. Steer, E. F. Smith, Z. R. Kudrynskiy, et al. 2D Mater. **5**, 035026(2018)

[3] K. S. Wickramasinghe, C. Forrester, M. C. Tamargo. Crystals. **13**, 677(2023)