

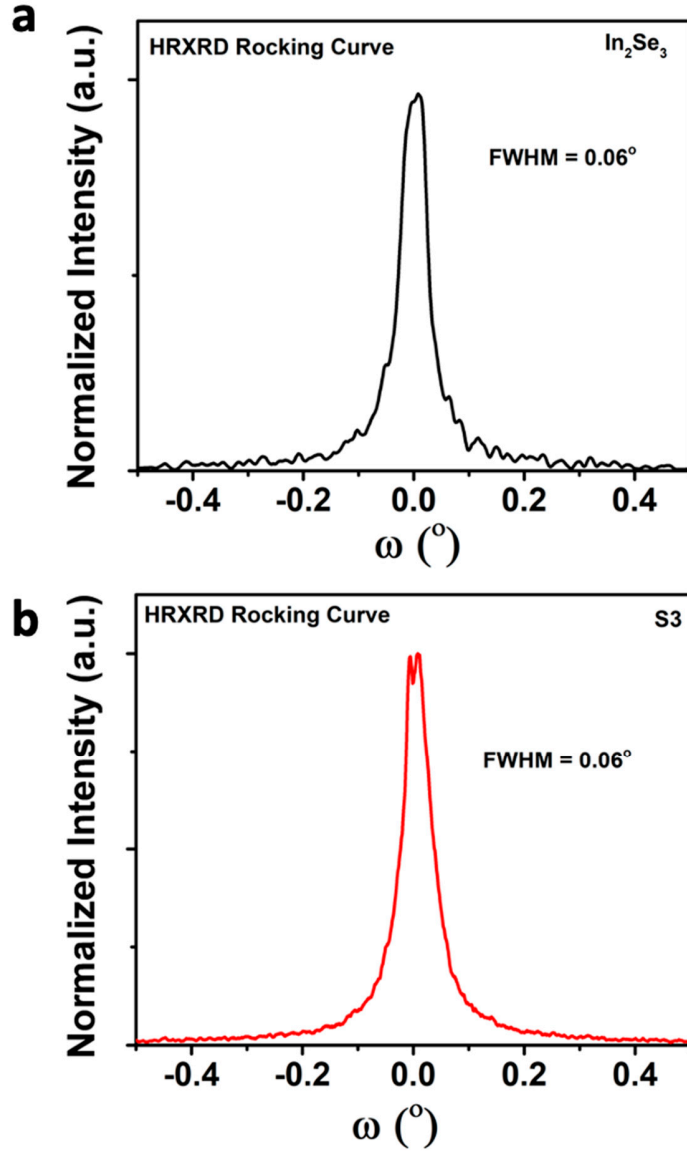
Molecular Beam Epitaxy of Twin-Free Bi₂Se₃ and Sb₂Te₃ on In₂Se₃/InP(111)B Virtual Substrates

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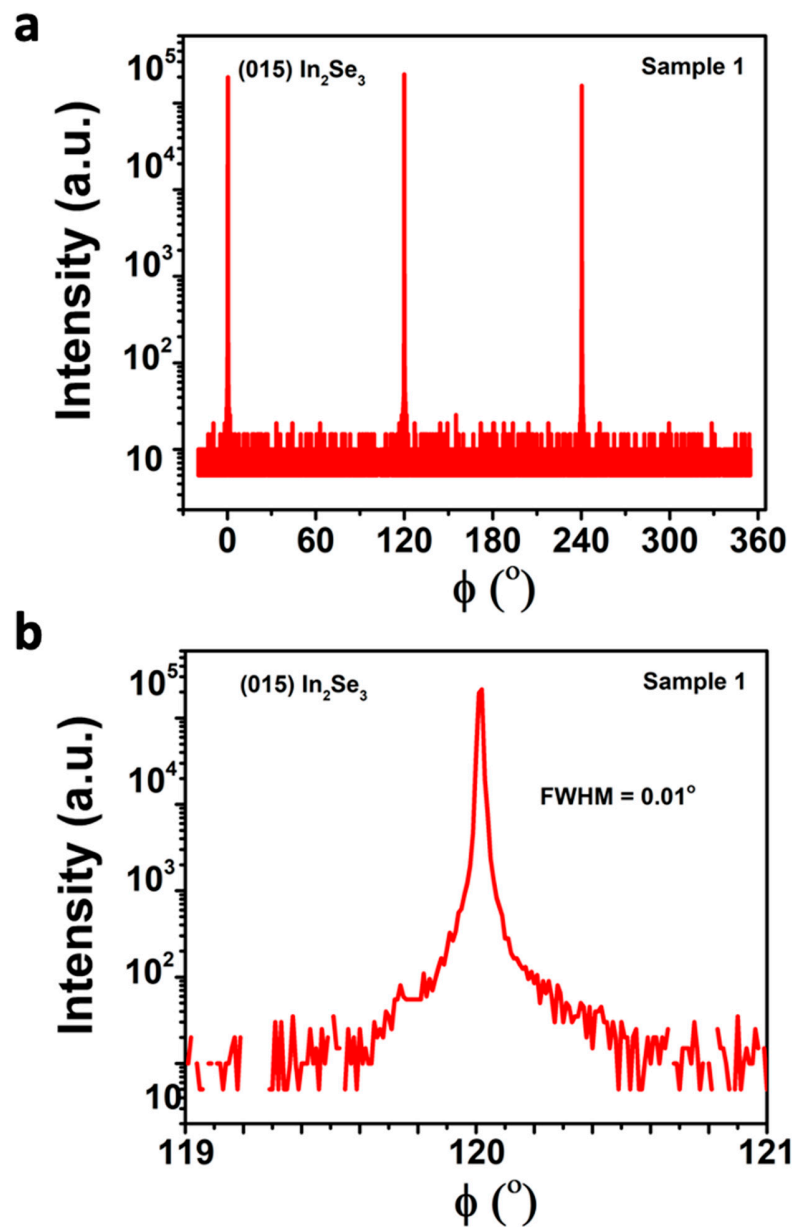
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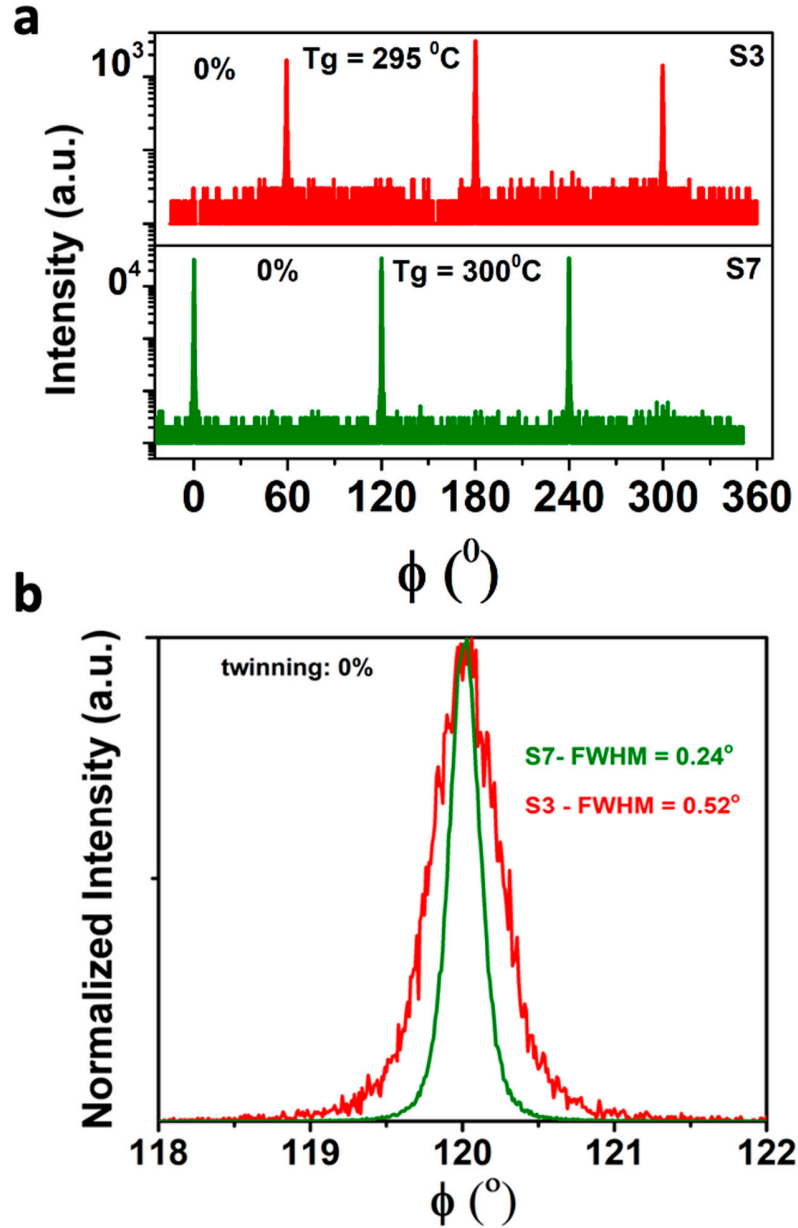
Supplementary Information



Supplementary Figure S1. High resolution X-ray diffraction rocking curve (RC) of (006) plane of: (a) 6 nm thick In₂Se₃ layer on InP(111)B substrate (i.e., In₂Se₃ virtual substrate) with a full width at half maximum (FWHM) of 0.06 $^{\circ}$ and (b) 18 nm thick Bi₂Se₃ layer (sample S3) grown on an In₂Se₃ virtual substrate with a FWHM of 0.06 $^{\circ}$. Bragg reflection angle (2θ) of (006) In₂Se₃ and (006) Bi₂Se₃ are 18.863 $^{\circ}$ and 18.559 $^{\circ}$, respectively.



Supplementary Figure S2. (a) ϕ scan of (015) plane of the un-twinned In_2Se_3 Sample 1. (b) Expanded view of the peak at 120° shown in (a) with a FWHM of 0.01° .



SUPPLEMENTARY FIG. 3. (a) ϕ scans of (015) plane of the fully twin suppressed Bi_2Se_3 samples S3 (18 nm thick) and S7 (64 nm thick). (b) Expanded view of the middle peak of the triplet shown in (a). The middle peak of S3 shown in (b) is shifted -60° to align with the middle peak of S7 for the convenience of comparison.