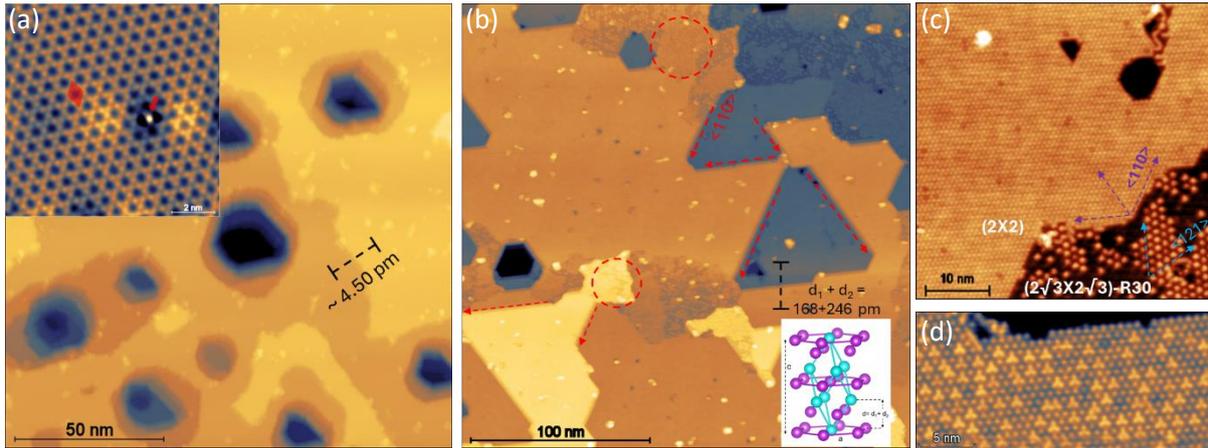


## Supplementary Information for

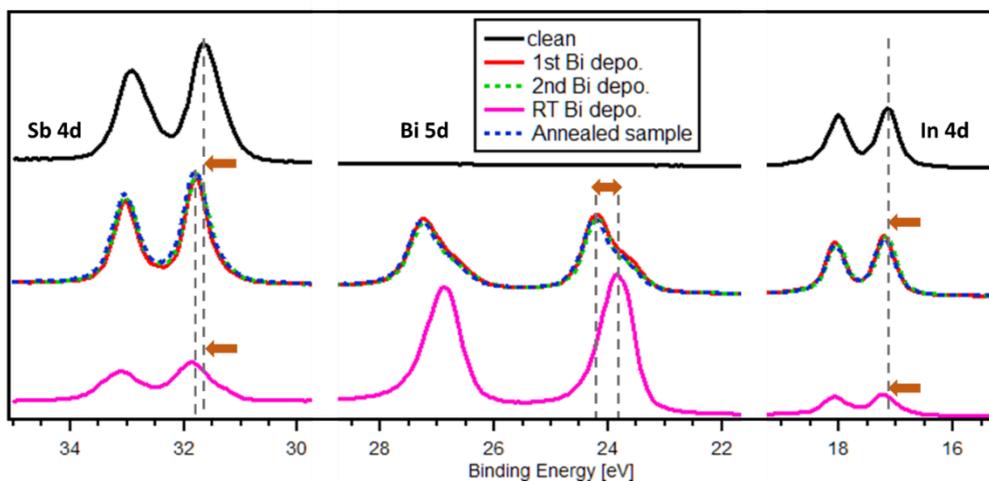
### **Bismuth-Trimer Adlayer and Thin Film Growth on In- and Sb-Terminated InSb(111) Surfaces**

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**Figure 1:** STM image of (a) oxide cleaned InSb(111)A showing InSb islands (inset shows atomic resolution of In-trimers, unit cell highlighted in red); (b) Hexagonal Bi(111) terrace on InSb(111)A, measured height constants matches with Bi-bilayer thickness (shown in the Bi(111) crystal structure); (c) STM image at the interface shows two types of reconstruction i.e. (2x2) and (2√3X2√3)-R30° along different symmetry directions; (d) Bias dependent STM image highlighting decorated Bi-trimers on InSb(111)A in the  $V_{Bias}$  range = +1.5 V to -1.5 V.



**Figure 2:** Combined XPS spectra of Bi/InSb(111)B, comparing Sb4d, Bi5d, and In4d core-level for clean and multiple cycles of Bi-deposition. Bi5d core-level shows the self-limiting nature of Bi incorporation at elevated sample temp. (marked as 1st and 2nd Bi depo. in the graph). RT Bi depo. shows dominating metallic Bi in the Bi5d spectrum. The reversibility of Bi incorporation can be seen in the Bi5d spectrum after annealing of the RT Bi deposited sample (marked as the Annealed sample in the graph). High B.E. shoulder component to Bi5d indicates mainly Bi-Sb bonding, further confirmed from peak fitting using the Voigt function.