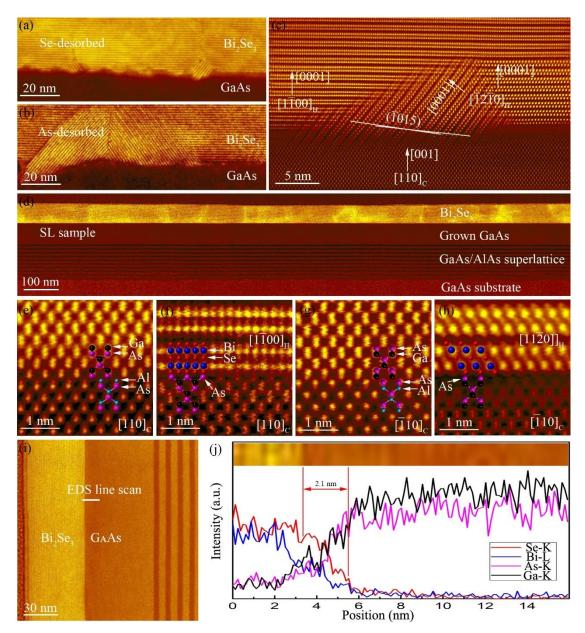
## **Supplementary Pages**



**Figure 1**. Cross-sectional HAADF-STEM characterization. (a, b) HAADF-STEM images taken along the GaAs  $[110]_{C}$  zone-axis showing the rough interface of Bi<sub>2</sub>Se<sub>3</sub>/GaAs in the Se-desorbed and As-desorbed samples, respectively. (c) Zoom-in atomic resolution image showing (0001) <sub>H</sub> oriented growth of  $[1\overline{100}]_{H}$  and  $[\overline{1210}]_{H}$  Bi<sub>2</sub>Se<sub>3</sub> domains on flat GaAs (001)<sub>C</sub> substrate, and  $(\overline{1015})_{H}$  oriented growth of  $[\overline{1210}]_{H}$  Bi<sub>2</sub>Se<sub>3</sub> domain on multi-faceted rough GaAs substrate. (d) Large-scale image demonstrating the flat interface of Bi<sub>2</sub>Se<sub>3</sub>/GaAs in the SL sample. (e-h) Zoom-in images overlapped with atomic models showing the

dumbbell atomic structure of the GaAs/AlAs superlattice in (e, g), and the atomically smooth interface of  $Bi_2Se_3/GaAs$  terminated by As-site (f, h) in the GaAs surface, respectively. (i) Rotated image showing the STEM-EDS line-scan analysis region. (j) Line-profiles of chemical compositions across the  $Bi_2Se_3/GaAs$  interface. False colors are added in the images to aid the eye.