

Figure 1 – Core level XPS on 1.5 nm Mn metal overlayer on  $Bi_2Se_3$  demonstrating a ~1.7 eV reduction in binding energy in Bi core levels compared to pristine  $Bi_2Se_3$ . Se migrates from below  $Bi_2Se_3$  surface into the growing Mn metal.



Figure 2 – (a) Core level XPS on 1 ML  $MnSe_x$  overlayer on  $Bi_2Se_3$  showing a rigid 0.8 eV shift toward higher binding energy in all  $Bi_2Se_3$  core levels. (b) STM height maps showing variation across the sample surface in monolayer thickness due to formation of  $\alpha$ -MnSe vs. MnSe<sub>2</sub>



Figure 3 – Schematic view of expected interface between  $\alpha$ -MnSe and Bi<sub>2</sub>Se<sub>3</sub>. Negatively charged sheets of Se anions sit above positively charged Mn cations creating dipole at the sample surface