Supporting Information

Molecular Beam Epitaxy of Two-Dimensional In_2Se_3 and its Giant Electroresistance Switching in Ferroresistive Memory Junction

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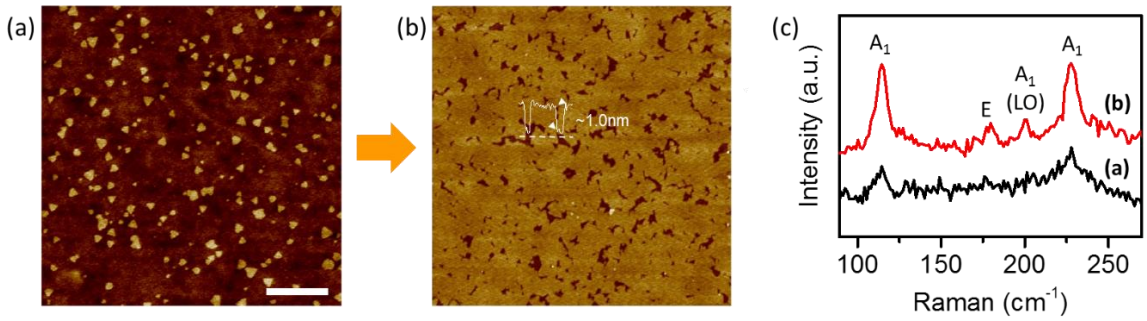

Figure S1. MBE growth of monolayer InSe. AFM topography of (a) the initial growth of InSe and (b) the subsequent coalescence to form a continuous monolayer. (c) Raman spectra of the respectively InSe grown at the different stages shown in (a) and (b). Scale bar for both AFM images: 600 nm.

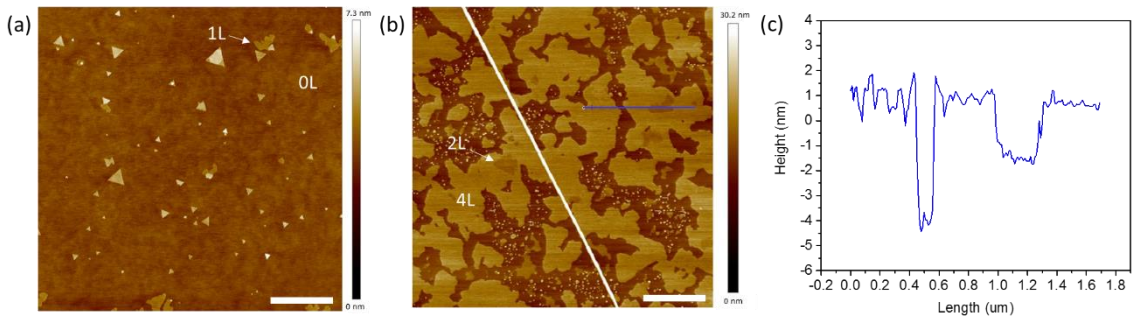

Figure S2. Desorption of grown α-In_2Se_3 at high temperature. (a) AFM topography of α-In_2Se_3 grown at 300 ^{0}C for 5 hrs. Despite the long hours, very scarce growth is observed. It is noted that growth at temperature higher than 300 ^{0}C for >5 hrs shows no visible growth. (b) AFM topography of 250^{0}C-grown 4L of α-In_2Se_3 after annealing at 400 ^{0}C for 1hr. The fully covered film desorbed to reveal substrate surface, as well as pits of 2L deep, as shown by (c) the height profile of the blue line in (b). The white line across is a wrinkle on graphene substrate. Scale bar: (a) 1.5 um, (b) 1um.

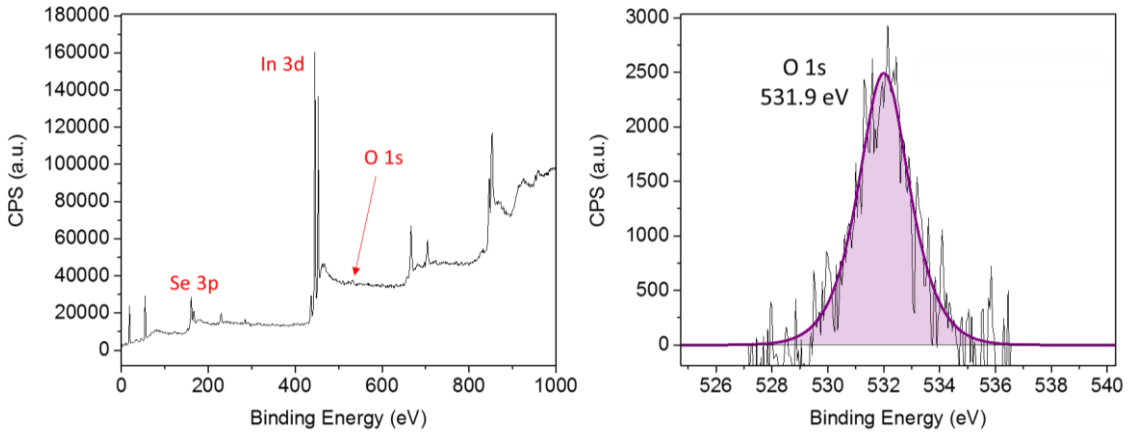

Figure S3. XPS of oxygen 1s core orbital peak. The position of the peak is found to be 531.9 eV, which is assigned to physisorbed oxygen. No oxide peak is detected at 530 eV. The amount of surface physisorbed oxygen is found to be very low, at 3.4%.

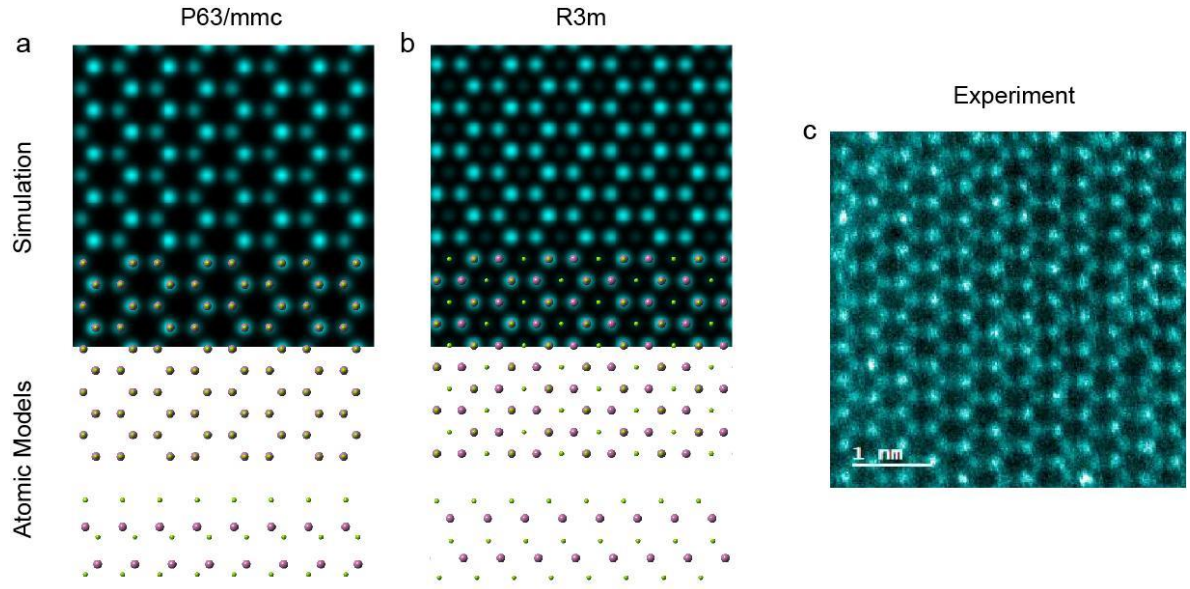

Figure S4. Identifying the structural phase of α-In_2Se_3. (a) Simulation of the *P63/mmc*[1] and (b) *R3m*[2] structural phases. Atomic models are overlaid, and the side views are depicted in the lower panels. (c) Atomic-resolution STEM-ADF image of the as-grown α-In_2Se_3 monolayer film, which indicates that it belongs to the *P63/mmc* space group.

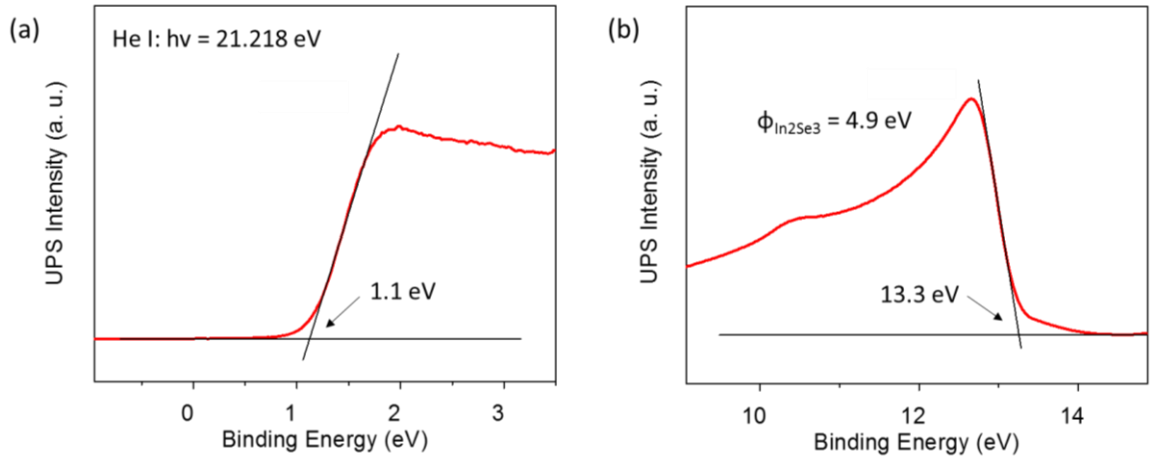

Figure S5. UPS of MBE grown α-In_2Se_3. (a) The valence band maximum (VBM) can be extracted by linearly extrapolating the leading edge of the spectrum to the baseline and was found to be 1.14 eV below the Fermi level. Using the electronic bandgap obtained using HREELS of 1.55 eV, the position of the Fermi level is found to be closer to the conduction band minimum (CBM) than the VBM, implying an intrinsic n-type doping of the sample. This is consistent with our device measurements. (b) The secondary electron cut-off edge is extracted by linearly extrapolating from the edge to the baseline and was found to be 13.3 eV. The work function is determined to be: 4.9 eV = 21.218 eV – 13.3 eV – 3 eV.

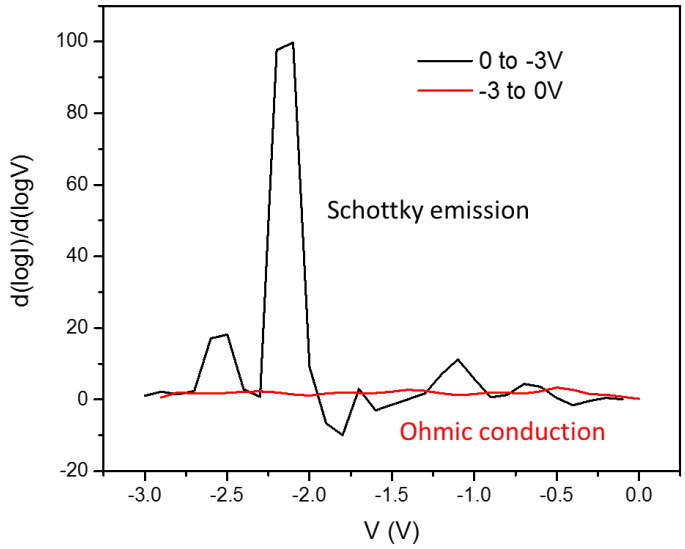

Figure S6. Conduction mechanism in MBE grown α-In_2Se_3. For the voltage sweep from -3 to 0 V (red), ohmic behavior is observed, in which dlog(I)/dlog(V) ≈ 1. For the voltage sweep from 0 to -3 V (black), increasing slope of log(I)-log(V) curve is observed, which indicates Schottky barrier controlled conduction.

References

1. Zhou, J.; Zeng, Q.; Lv, D.; Sun, L.; Niu, L.; Fu, W.; Liu, F.; Shen, Z.; Jin, C.; Liu, Z. *Nano Lett.* **2015,** 15, 6400-6405.
2. Xiao, J.; Zhu, H.; Wang, Y.; Feng, W.; Hu, Y.; Dasgupta, A.; Han, Y.; Wang, Y.; Muller, D. A.; Martin, L. W.; Hu, P.; Zhang, X. *Phys. Rev. Lett.* **2018,** 120, 227601.