Supporting information for

Vertically Stacked $Bi_2Se_3/MoTe_2$ Heterostructure with Large Band Offsets for Nanoelectronics

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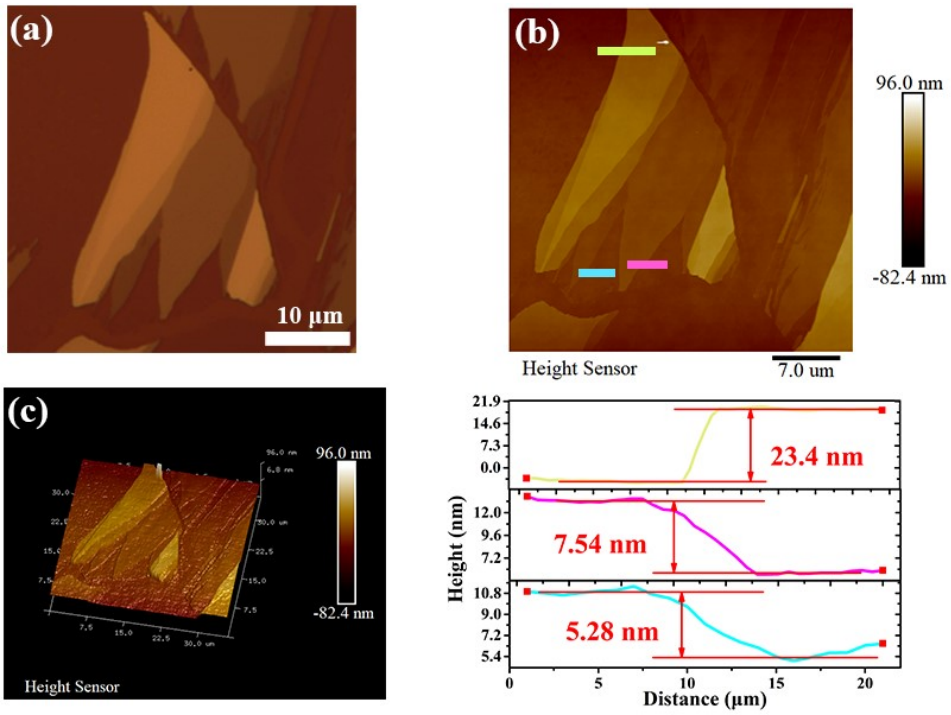

Fig. S1 Bi_2Se_3 nanosheets prepared by the Au-assisted exfoliation method. a Optical image of the Bi_2Se_3 nanosheets on Ti/Au SiO_2/Si substrate. **b** the 2D topological AFM image of Bi_2Se_3 nanosheets on Ti/Au SiO_2/Si substrate. **c** the corresponding 3D topological AFM image from **b**. **d** the height profiles along the yellow, blue and pink lines from **b**.

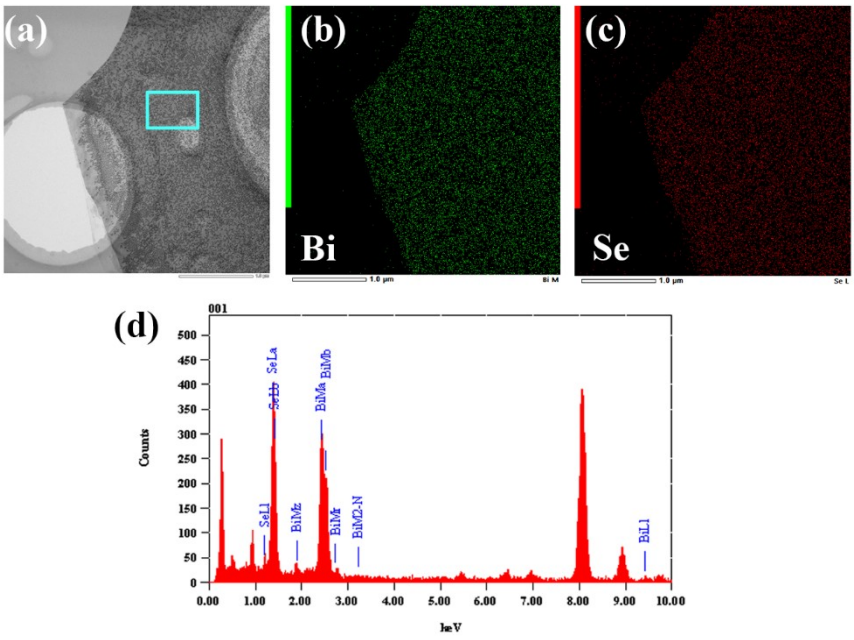

Fig. S2 HRTEM-EDS mapping analysis of the Bi_2Se_3/Au nanosheets. a Low-magnification TEM image of the exfoliated and as-transferred Bi_2Se_3 nanosheets with Au particle. **b** Element mapping of the two detected elements: Bi and **c** Se. **d** EDS spectrum extracted from the light blue rectangle in **a**.

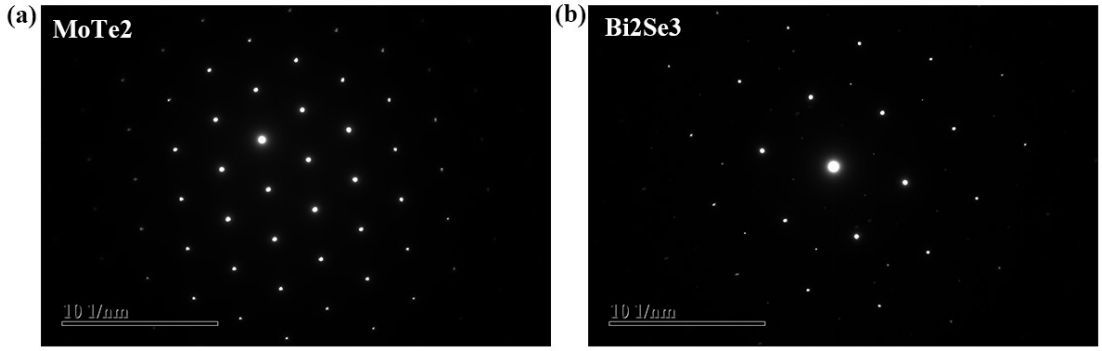

Fig. S3 SAED patterns of (a) individual $MoTe_2$ nanosheets and (b) individual Bi_2Se_3 nanosheets

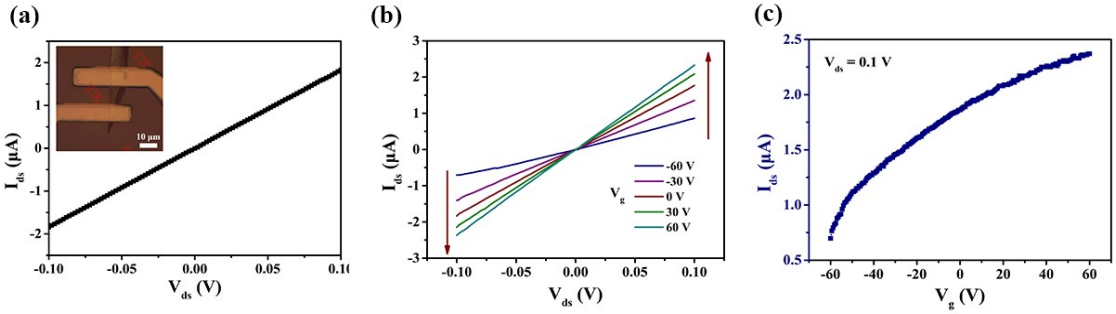

Fig. S4 Electrical performance of another ultrathin Bi_2Se_3 FET. a. I_{ds}-V_{ds} curve. **b.** Output curves at various gate voltages. **c.** Tranfer characteristic at V_{ds} = 0.1 V scanning from -60 V to 60 V

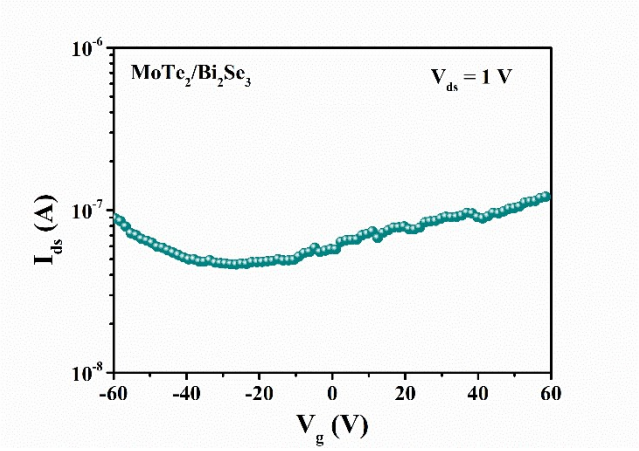

Fig. S5 Transfer curve of the heterojunction device I at V_{ds} = 1 V

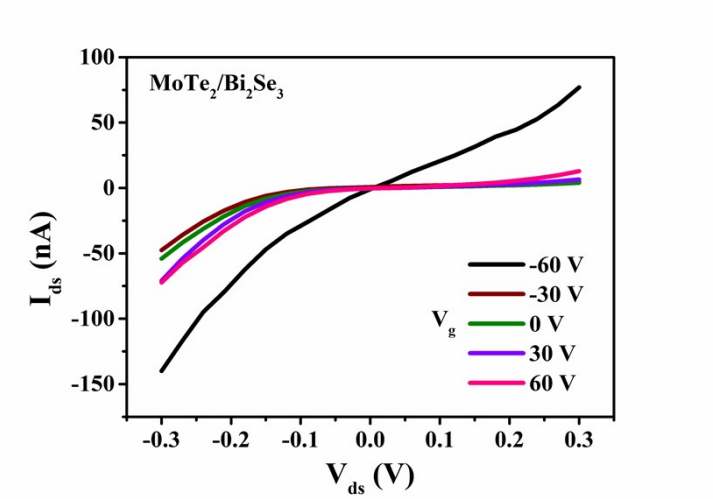

Fig. S6 Output curves of the heterojunction device I at various gate voltages

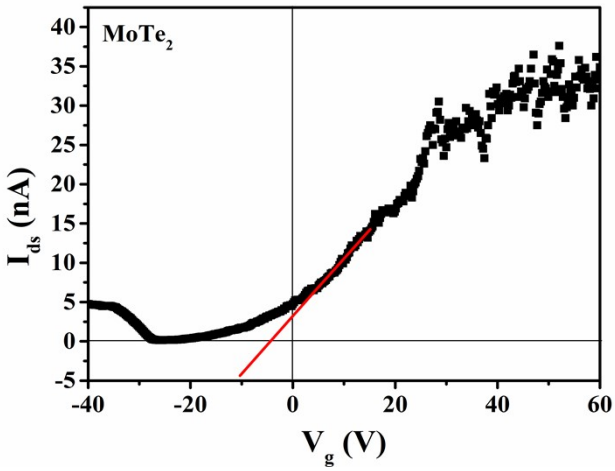

Fig. S7 Transfer curve of $MoTe_2$ FET in device I at V_g scanning from -40 V to 60 V. The threshold voltage (V_{th}) can be extracted from the intersection point between redfitting curve and the horizontal line.

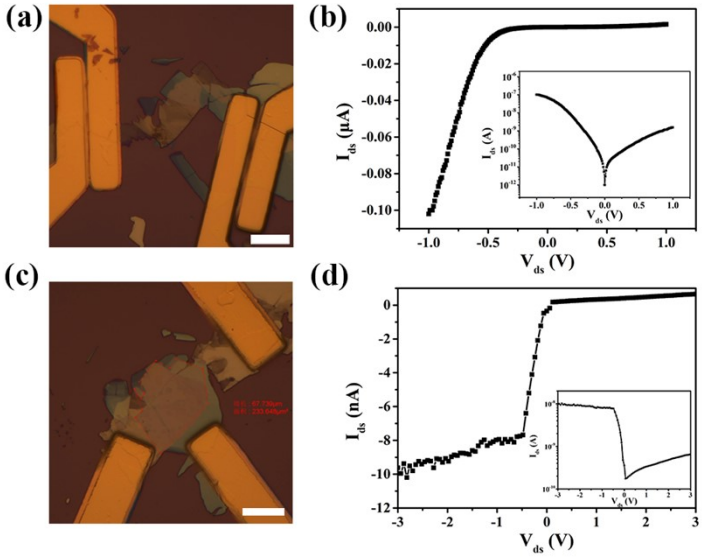

Fig. S8 Electrical properties of another two $MoTe_2/Bi_2Se_3$ heterostructure devices. (a) OM image of Device II, (b) the corresponding I_{ds}-V_{ds} curve at linear form, Inset is the logarithmic scale. (c) OM image of Device III, (d) the corresponding I_{ds}-V_{ds} curve at linear scale, Inset is the logarithmic scale.

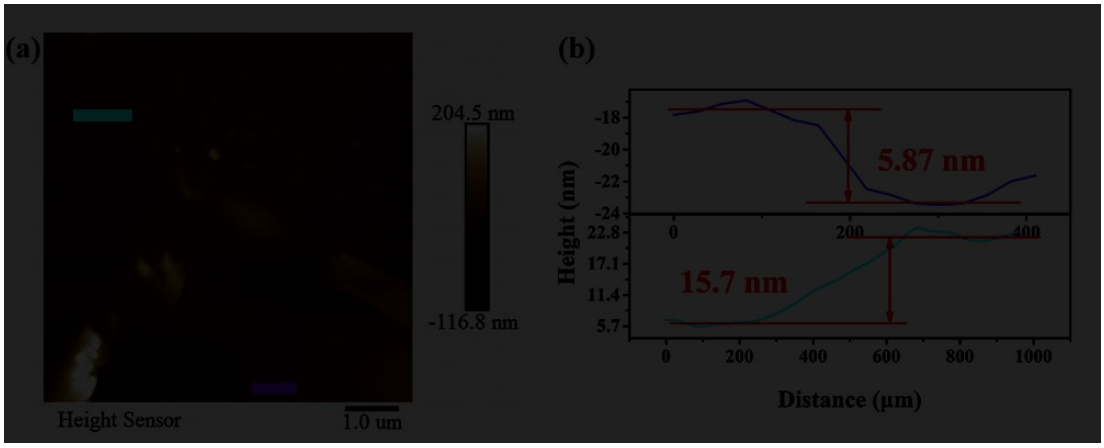

Fig. S9 (a) Topological AFM image of $Bi_2Se_3/MoTe_2$ in device VI. (b) the corresponding height profile along the cambrige blue and violet line, respectively.

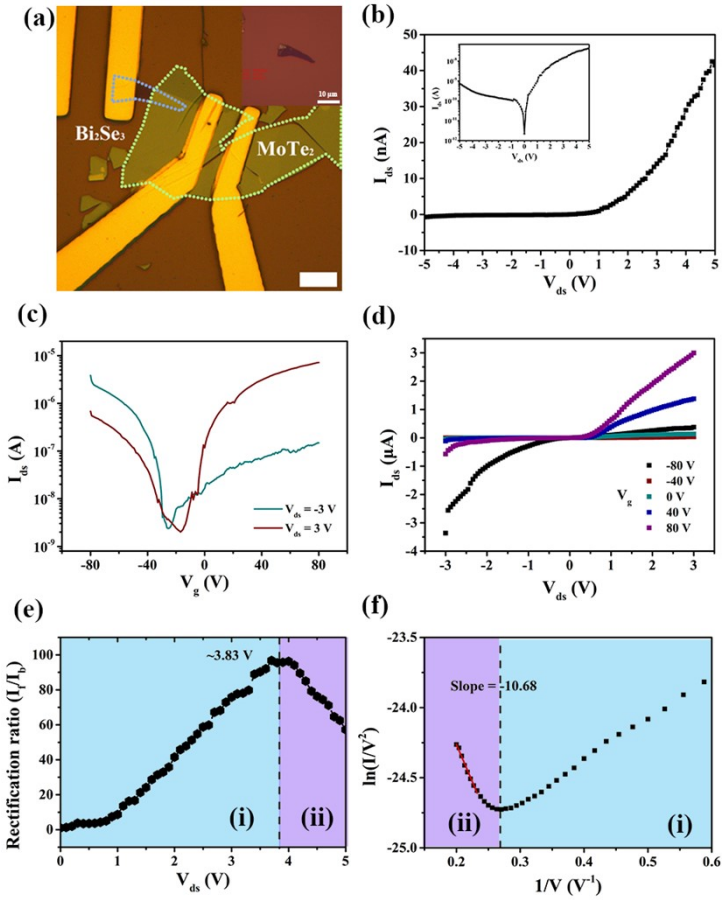

Fig. S10 Electrical properties of another $MoTe_2$/thinner Bi_2Se_3 heterostructure devices. (a) OM image of Device IV. The scale bar is 10 μm. Inset is the OM image of the chosen Bi_2Se_3 nanosheets. (b)I_{ds}-V_{ds} curve at linear scale. Inset is the semi-logarithmic scale. (c) Transfer curves at V_{ds} = -3 V/3 V, (d) Output curve at linear scale from V_g = -80 to 80 V. (e)rectification ratio (I_f/I_b) at the same V_{ds} with the increment of V_{ds}. (f) Fowler-Nordheim plot for backward current. Noticement: Bi_2Se_3 is Drain terminal and 2H-MoTe is Source terminal.

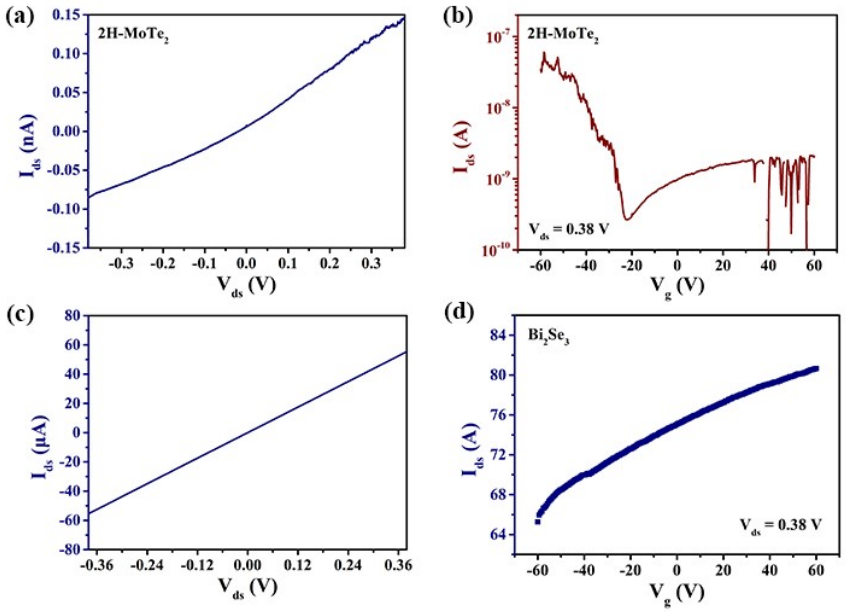

Fig. S11 Electrical properties of individual $MoTe_2$ and Bi_2Se_3 at V_{ds} = 0.38 V in device I. I_{ds}-V_{ds} curves for (a) $MoTe_2$ and (c) Bi_2Se_3; Transfer characteristics for (b) $MoTe_2$ and (d) Bi_2Se_3.

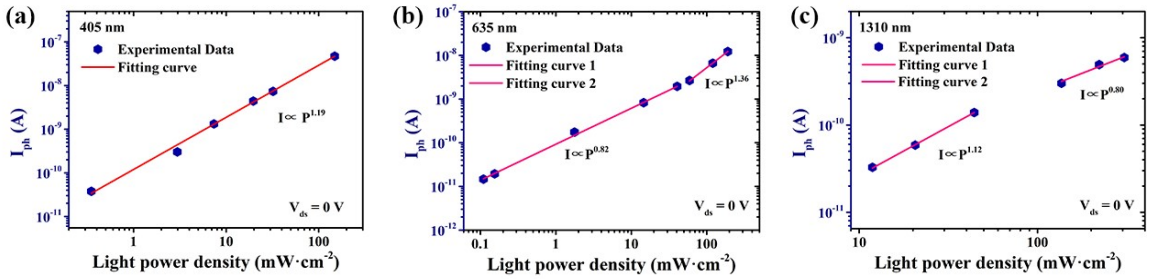

Fig. S12 The fitting relationship of the logarithmic plots between photocurrent and light power density under (a) 405 nm, (b) 635 nm and (c) 1310 nm illumination.

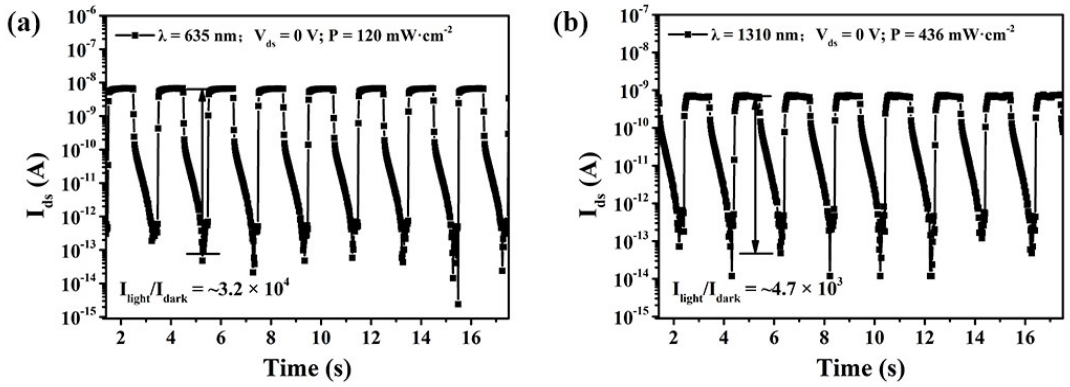

Fig. S13 Time trace of I_{ds} at V_{ds} = 0 V under (a) 635 nm, (b) 1310 nm illumination.

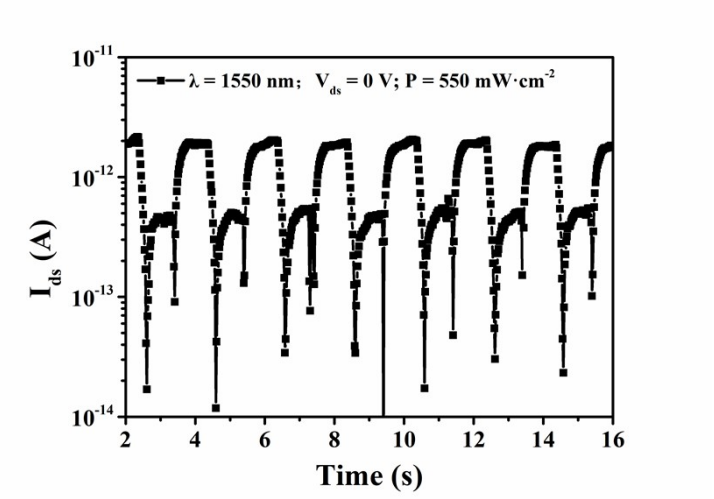

Fig. S14 Time trace of I_{ds} at V_{ds} = 0 V under 1550 nm illumination.

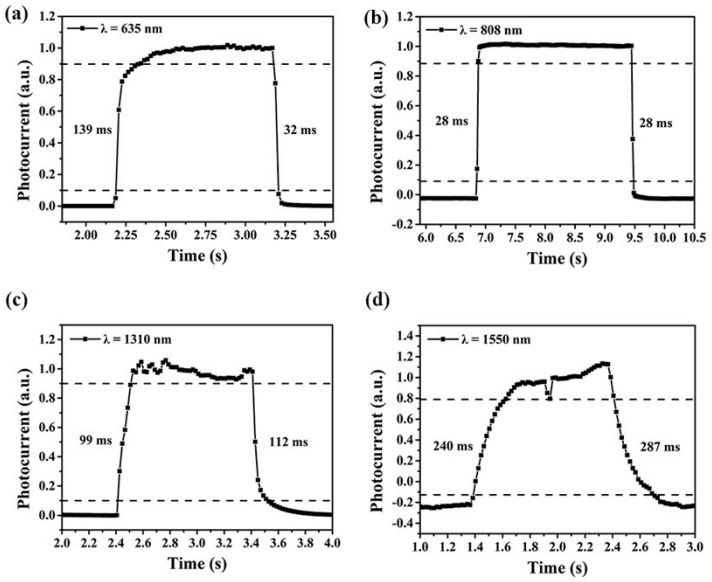

Fig. S15 The cooresponding rise and decay time of the device I at V_{ds} = 0 V under **a.** 635 nm (P = 120 mW/cm^2). **b.** 808 nm (P = 146 mW/cm^2). **c.** 1310 nm (P = 350 mW/cm^2) and **d.** 1550 nm (P = 550 mW/cm^2) illuminations.

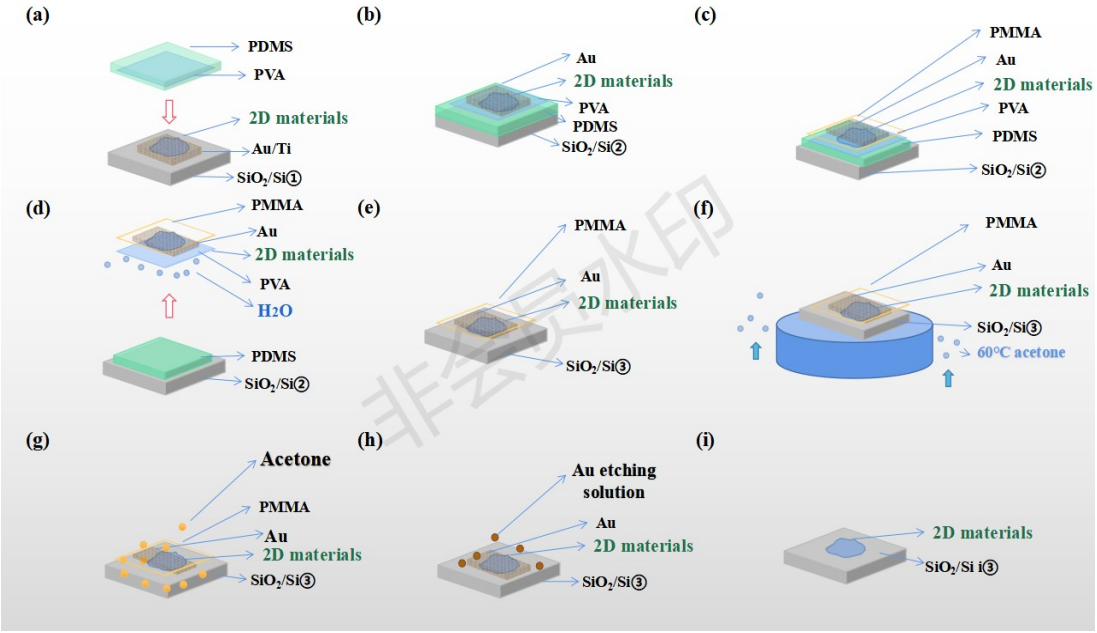

Fig. S16 3D schematic image of our novel exfoliation and transfer process for Bi_2Se_3 nanosheets.

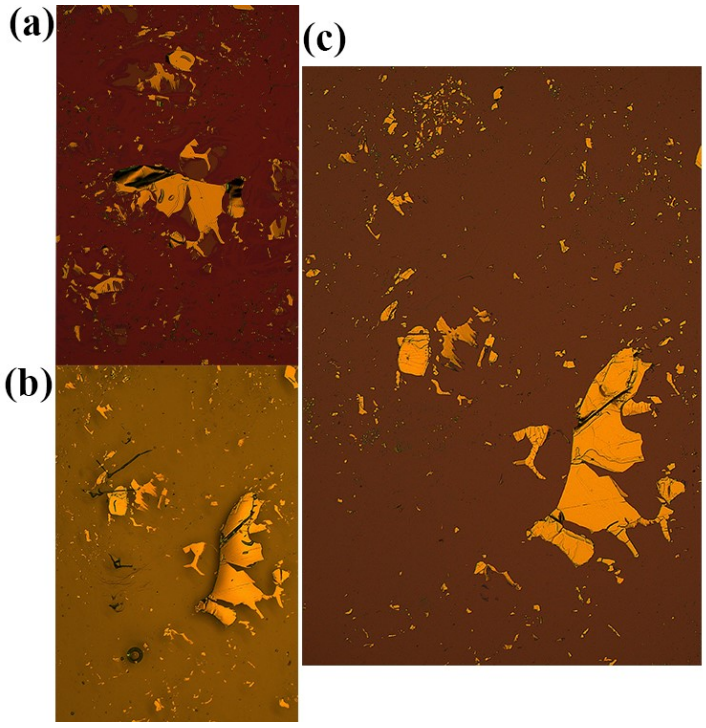

Fig. S17 Optical images of the Au-assisted exfoliation and transfer process: (a) large-scale Bi_2Se_3 samples on Au/Ti/SiO_2/Si substrate; (b) large-scale Bi_2Se_3 samples on PVA/PDMS film; (c) large-scale Bi_2Se_3 samples on final SiO_2/Si substrate.

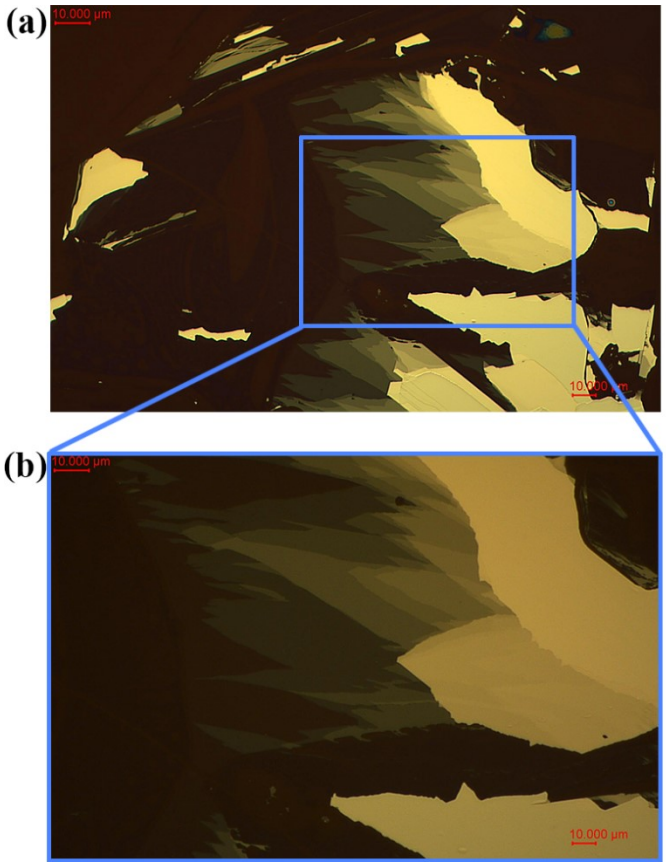

Fig. S18 Ultra-large Bi_2Se_3 nanosheets on Au/Ti/SiO_2/Si substrate at (a) 10 X and 50 X amplification.

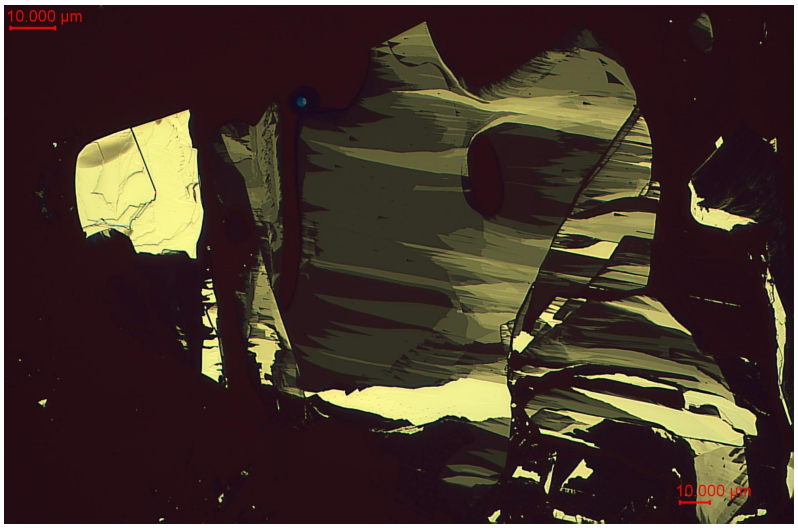

Fig. S19 Another ultra-large Bi_2Se_3 nanosheets on Au/Ti/SiO_2/Si substrate at 20 X amplification.