

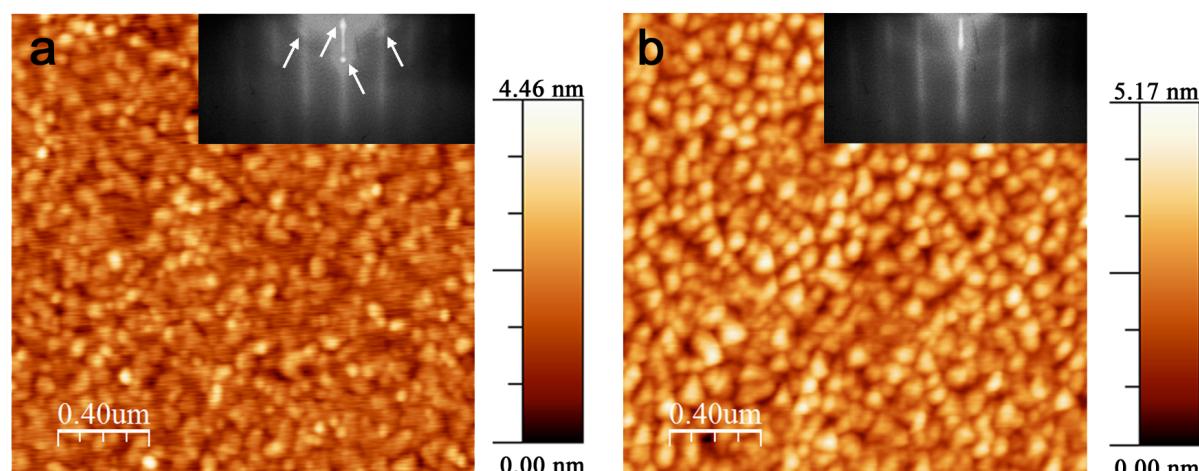
## Supplementary Information

### Ordered Growth of Topological Insulator $\text{Bi}_2\text{Se}_3$ Thin Films on Dielectric amorphous $\text{SiO}_2$ by MBE

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#### 1. The influence of Se passivation

Fig. S1a and b show AFM images of  $\text{Bi}_2\text{Se}_3$  films grown on unpassivated  $\text{SiO}_2$  surface and Se passivated  $\text{SiO}_2$  surface, respectively. Surface morphology is obtained by commercial AFM (NanoFocus Inc). Both of samples were not annealed after growth of 2 hours. The morphology of as-grown films without passivation is exhibited small grains (Fig. S1a) and RHEED pattern was indicated a formation of other phase as shown in inset of Fig. S1a (marked by white arrow). On the other hand, as-grown films on Se passivated  $\text{SiO}_2$  presents forming of large grains (Fig. S1b) and RHEED pattern of well-crystallized  $\text{Bi}_2\text{Se}_3$  (the inset of Fig. S1b). The surface roughness is 0.54 nm and 0.66 nm for  $\text{Bi}_2\text{Se}_3$  films grown on the unpassivated surface and the passivated surface, respectively.

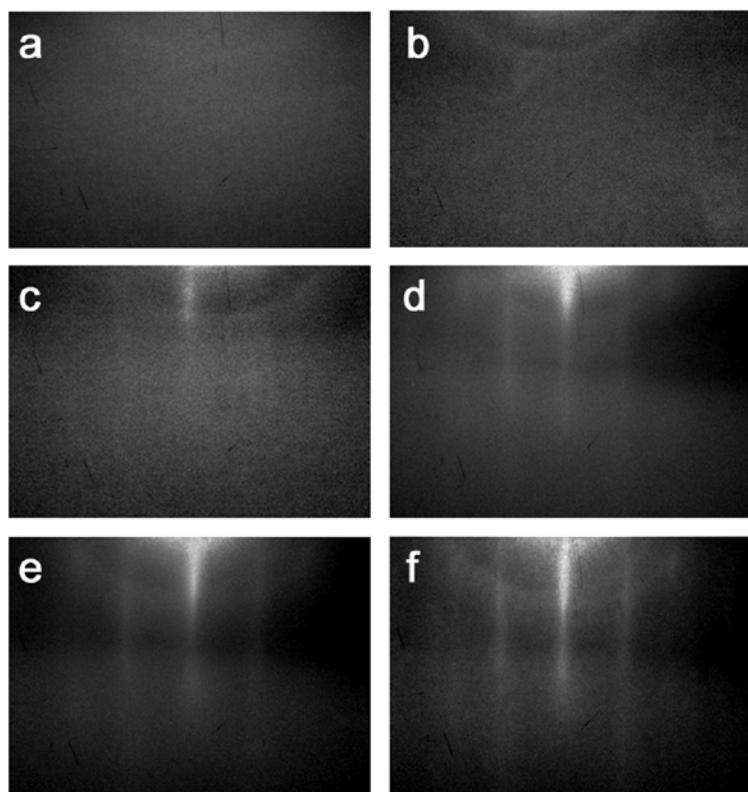


**Fig. S1** AFM images of  $\text{Bi}_2\text{Se}_3$  films for (a) grown on unpassivated  $\text{SiO}_2$  surface and (b)

Se passivated  $\text{SiO}_2$  surface. The insets are RHEED patterns recorded during the growth.

## 2. RHEED patterns for $\text{Bi}_2\text{Se}_3$ growth procedures

Prior to the growth, samples were heated at 120 °C to remove residual contamination. Fig. S2a shows RHEED pattern of  $\text{SiO}_2$  substrate at 120 °C, which is typical RHEED pattern for amorphous substrate. After 30 minutes, Se was deposited on  $\text{SiO}_2$  surface. RHEED pattern was not changed from that of amorphous (Fig. S2b). Then, temperature increased to 250 °C for growth of  $\text{Bi}_2\text{Se}_3$  films. During the growth, RHEED pattern is developed to streaky diffraction pattern as shown in Fig. S2c and S2d. After the growth, samples were annealed at 450 °C (Fig. S2e). RHEED patterns are not shown the formation of other phase in the annealing to cool down (Fig. S2f).

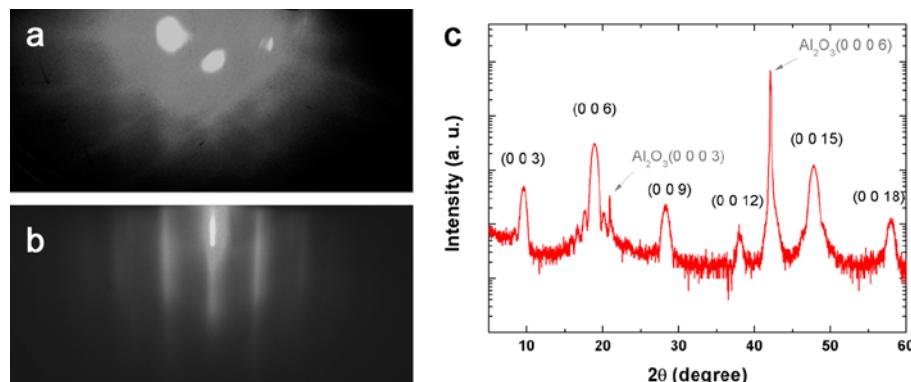


**Fig. S2** RHEED patterns for  $\text{Bi}_2\text{Se}_3$  growth procedures: (a)  $\text{SiO}_2$  substrate at 120 °C, (b) Se exposure for passivation, (c)-(d)  $\text{Bi}_2\text{Se}_3$  growth (RHEED recorded at 30 min and 90 min,

respectively), (e) annealing at 450 °C for 30 min, and (f) cool down. The large dark features on the screen are shadows of the substrate holders.

### 3. RHEED pattern and XRD profile of epitaxially grown Bi<sub>2</sub>Se<sub>3</sub> films on Al<sub>2</sub>O<sub>3</sub>(0001) substrate

Fig. S3 provides RHEED patterns and XRD profile of MBE-grown epitaxial Bi<sub>2</sub>Se<sub>3</sub> films on Al<sub>2</sub>O<sub>3</sub>(0001), known as sapphire, substrate. As shown in Figure S3a, RHEED pattern indicates crystalline sapphire substrate. During the growth of Bi<sub>2</sub>Se<sub>3</sub> at 250 °C, RHEED pattern changes to the crystalline Bi<sub>2</sub>Se<sub>3</sub>. XRD profile, obtained by Shimadzu XRD-6100, reveals the (003) family of diffraction peak of c-axis oriented Bi<sub>2</sub>Se<sub>3</sub> films as shown in Fig. S3c.

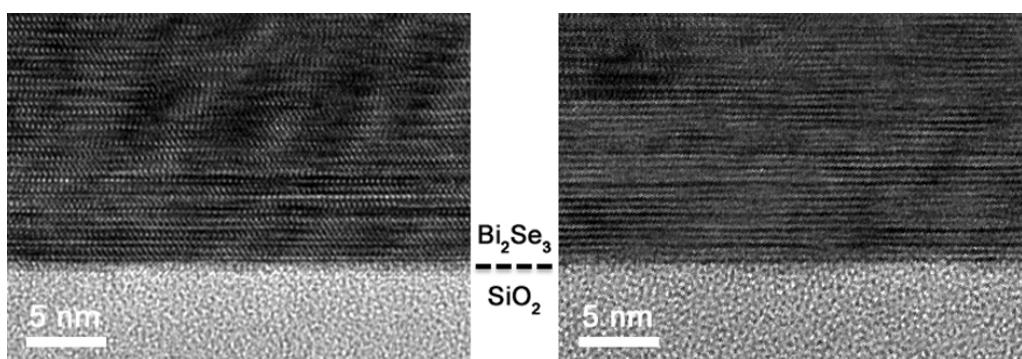


**Fig. S3** RHEED pattern of (a) crystalline sapphire substrate and (b) epitaxially grown Bi<sub>2</sub>Se<sub>3</sub> films. (c) XRD profile of MBE-grown Bi<sub>2</sub>Se<sub>3</sub> films on sapphire substrate.

### 4. Additional HRTEM images of interface between Bi<sub>2</sub>Se<sub>3</sub> films and flat SiO<sub>2</sub>

Fig. S4 shows the HRTEM images for different Bi<sub>2</sub>Se<sub>3</sub> thin film samples grown on SiO<sub>2</sub>. Additional HRTEM measurement was carried out using JEM-2100F instrument operated at

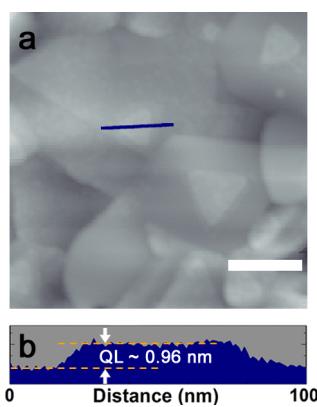
200 kV. Well-stacked  $\text{Bi}_2\text{Se}_3$  quintuple layers are clearly observed.



**Fig. S4** HRTEM images for  $\text{Bi}_2\text{Se}_3$  thin films grown on flat  $\text{SiO}_2$  surface area.

## 5. Surface morphology of $\text{Bi}_2\text{Se}_3$ films grown on $\text{SiO}_2$

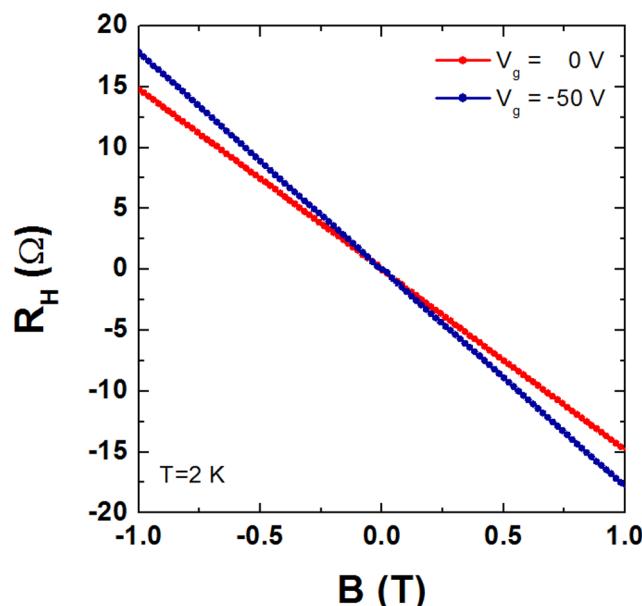
Fig. S5 shows the AFM image of a 20 nm thick  $\text{Bi}_2\text{Se}_3$  thin film grown on  $\text{SiO}_2$ . Terraces and/or steps (with triangular and hexagonal shapes) represent the 3-fold symmetry of  $\text{Bi}_2\text{Se}_3$ . The cross-sectional height of triangular feature is  $\sim 0.96$  nm. The disordered feature of triangular  $\text{Bi}_2\text{Se}_3$  terraces along the in-plane directions can be attributed to the 300 nm thick amorphous  $\text{SiO}_2$  surface which does not provide a lattice constraint for atom stacking during the deposition.



**Fig. S5** (a) AFM image of a 20 nm thick film. Scale bar = 100 nm. (b) Cross-sectional profile along the line in (a) showing the QL of  $\text{Bi}_2\text{Se}_3$ .

## 6. Hall measurement for 20 nm thick films

Fig. S6 shows Hall resistance of 20 nm thick films in the perpendicular magnetic field. The standard six-probe method is used for Hall measurement ( $I = 100 \mu\text{A}$ ,  $T = 2 \text{ K}$ ). Sample is indicated as n-type semiconductor with a carrier concentration of  $4.2 \times 10^{13} \text{ cm}^{-2}$  at  $V_g = 0 \text{ V}$ . The carrier density decreased down to  $3.4 \times 10^{13} \text{ cm}^{-2}$  at  $V_g = -50 \text{ V}$ .



**Fig. S6** Hall resistance of 20 nm thick films for applying gate voltage in the magnetic field of  $\pm 1 \text{ T}$ .