Supplementary Pages
(Ultra-thin Bi_2Se_3 Films Grown by Molecular Beam Epitaxy)

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Figure S1: The $5 \times 5\ \mu m$ AFM images of the 4 nm thin Bi_2Se_3 grown on sapphire (0001) substrate using two-step method directly. The grown film is not continuous as we can see many dips in the AFM and the rms surface roughness of the sample is very high.

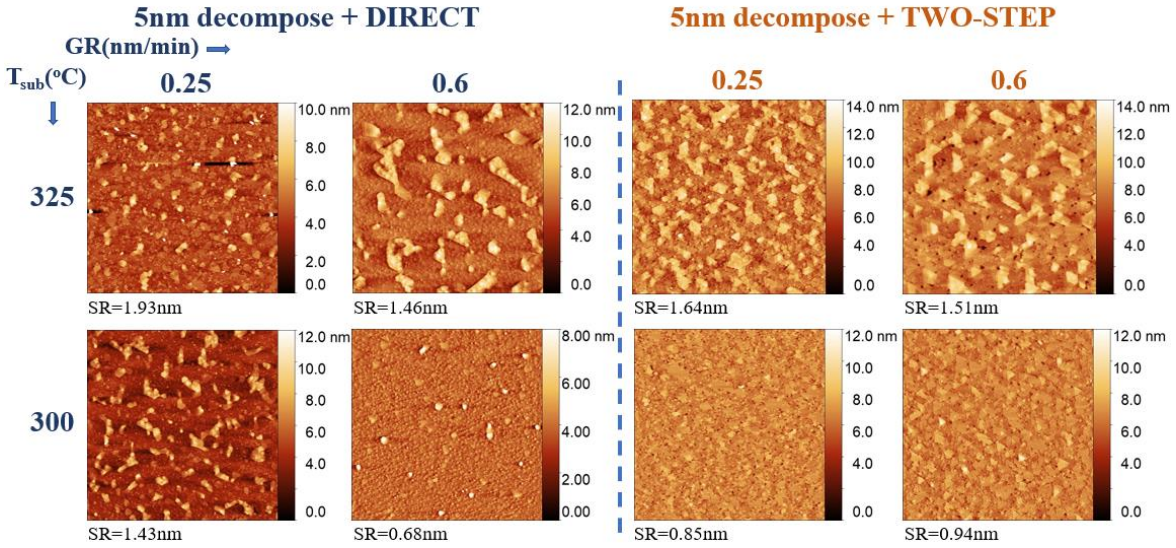

Figure S1: The $5 \times 5\ \mu m$ AFM images of the 4 nm thin BS films deposited by direct and two-step growth methods after the decomposition of 5nm BS. The growth rate and the surface

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temperatures are mentioned on the top and left for respective columns and rows. The second growth temperature is 100^{o}C higher than the mentioned substrate temperature for two-step growth. The root-mean-square roughness (SR) is mentioned under each AFM accordingly.

GROWTH DETAILS

The sapphire (0001) substrates were heated at 200^{o}C in the load lock for 12 hours. Then they were transferred to the growth chamber where they were heated to 650^{o}C and kept there for 5 minutes to desorb impurities. Then they were cooled down to the desired growth temperature for depositing the thin films or surface treatment.

Surface treatment: To change the surface energy 5nm of Bi_2Se_3 (BS) was grown directly on the substrates at 325^{o}C. Then we heated the substrates with continual Se flux to 470^{o}C, which is higher than the BS decomposition temperature for our MBE. We kept the substrates there for 30 minutes to completely desorb the grown BS. Then the substrates were cooled to the required temperatures for growth using direct or two-step method.

DIRECT GROWTH: For direct growth, we opened the bismuth and selenium shutter together for the ions to travel and get deposited on the rotating substrate.

TWO-STEP GROWTH: 3 nm of the thin film was grown using the grow anneal strategy at the first growth temperature, where we grew BS for a minute with co-deposition of bismuth and selenium and annealed for eighty seconds with continual Se. Then we raised the substrate temperature to the second set point (100^{o}C higher than the first growth temperature) under Se flux to grow the remaining 1nm film.

After the film was deposited the substrate was cooled down to 200^{o}C under the Se flux and moved to the load lock.

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