In situ reflection electron microscopy for investigation of surface processes on Bi_2Se_3(0001) during adsorption, sublimation, and growth

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Nanostructures based on layered 2D metal chalcogenides have proved their performance as infrared photodetectors [1]. To grow high-quality heterostructures and superlattices, molecular beam epitaxy is used, but various defects are generated during the growth: point defects [2], twin domain boundaries [3], antiphase boundaries caused substrate's atomic steps [4]. This puts a demand for *in situ* microscopy investigation of surface processes and growth mechanisms [5].

We used *in situ* reflection electron microscopy (REM) and *ex situ* atomic force microscopy (AFM) to study sublimation and van der Waals (vdW) epitaxy on the Bi_2Se_3(0001) surface. Using probe lithography, Bi_2Se_3(0001) surface was patterned with 3–15 nm deep grooves running along $\langle 11\bar{2}0\rangle$-type directions. Heating to ~400°C triggers ascending motion of atomic steps on the Bi_2Se_3(0001) surface that manifests the onset congruent sublimation observed first by *in situ* REM [6]. In this process, the grooves made on the Bi_2Se_3(0001) surface act as sources of atomic steps: groove depth increases, generates atomic steps, and they move in the ascending direction away from the source. We used this phenomenon to create self-organized regularly-spaced zigzag atomic steps having 1 nm height on the Bi_2Se_3(0001) surface.

Using *in situ* REM, we have studied transformation of the Bi_2Se_3(0001) surface, when a metal (Bi, In, or Sn) is deposited simultaneously with Se. When we started Bi deposition with rates up to ~0.01 nm/s at 200–400°C and constant Se flux (up to ~0.1 nm/s), there begins descending motion of atomic steps. This corresponds to the onset of Bi_2Se_3(0001) vdW epitaxy visualized first by *in situ* REM [6]. The deposition of In onto the Bi_2Se_3(0001) surface at ~400°C leads to nucleation of impurity-induced surface phase at step edges followed by its expansion on terraces associated with the increase of In coverage. The vdW heteroepitaxy started with 2D island nucleation and, after 3–5 nm growth, continued with the formation of 3D islands. We have not detected the formation of any impurity-induced phase during the Sn deposition, and layer-by-layer growth of $SnSe_2$ starts with the nucleation of 2D islands.

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References
[1] Wang F et al. 2D Metal Chalcogenides for IR Photodetection. Small. 2019;15:1901347.
[2] Callaert C et al. Interstitial defects in the van der Waals gap of Bi_2Se_3. Acta Crystallogr Sect B Struct Sci Cryst Eng Mater. 2019;75:717–732.
[3] Kampmeier J et al. Suppressing Twin Domains in Molecular Beam Epitaxy Grown Bi_2Te_3 Topological Insulator Thin Films. Cryst Growth Des. 2015;15:390–394.
[4] Borisova S et al. Domain formation due to surface steps in topological insulator Bi_2Te_3 thin films grown on Si (111) by molecular beam epitaxy. Appl Phys Lett. 2013;103:081902.
[5] Yang Z et al. Recent Progress in 2D Layered III–VI Semiconductors and their Heterostructures for Optoelectronic Device Applications. Adv Mater Technol. 2019;4:1900108.
[6] Ponomarev S et al. In situ reflection electron microscopy for investigation of surface processes on Bi_2Se_3(0001). J Phys Conf Ser. 2021;1984:012016.