Since the theoretical prediction [1] of surface states in Bi\(_2\)Se\(_3\) and their subsequent experimental confirmation by means of ARPES measurements [2], there have been no transport measurements unambiguously showing the presence of surface conductivity in Bi\(_2\)Se\(_3\). Defects like twinning, Se vacancies, mosaicity twist and tilt, are known to influence transport properties.

The goal of the present work was to reveal the origin of the formation of different structural defects in Bi\(_2\)Se\(_3\) thin films. We conducted a detailed study of layers grown by MBE on InP(111)A and -B terminated flat and rough substrates. This choice of substrate reduces the formation of mosaicity twist sufficiently due to an almost perfect lattice match (0.2%) between InP and Bi\(_2\)Se\(_3\).

Bismuth selenide layers grown on flat InP(111)B were found to have a "poor-crystalline quality" interface layer, which consists of crystalline domains with a thickness of one quintuple layer (QL). They are not always perfectly aligned to the substrate; misalignment occurs in areas where domains meet and try to merge. There are two difficulties in this process. First, the 'flat' InP(111)B substrate (R\(_{\text{RMS}}\) = 0.1 nm) is not atomically flat, but has diatomic surface steps with a height of 3.38 Å (0.35 QL). Second, even if two nucleation points form on a perfectly flat area, there is always a chance of twin formation, depending on how the second layer of the QL is formed (A−B−C or A−C−B). Because of both reasons we conclude that "2D information" passed on to the film by a flat substrate is not sufficient for realizing the controlled growth of Bi\(_2\)Se\(_3\).

AFM, XRD and STEM measurements of Bi\(_2\)Se\(_3\) grown on a rough InP(111)B substrate (R\(_{\text{RMS}}\) = 2.1 nm) reveal the absence of twin domains and a high-quality interface. Since the sides of the hollows are higher than the height of a QL (9.6 Å), they behave as additional \{1-11\} surfaces, so that both the substrate surface and the side surface of the hollow define the alignment of the QL layers and the stacking within a QL, providing the "3D information" that results in the unique layer stacking. Similar experiments performed using Fe-doped InP(111)A substrates showed the same tendency; the only difference between A and B terminated substrates was the particular family of twin domains that was suppressed by roughness. The suppression of twins results in a reduction of the carrier density up to 89% compared to values obtained for twinned Bi\(_2\)Se\(_3\) layers.


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Fig. 1: Cross-sectional HAADF-STEM images of the interface between a Bi$_2$Se$_3$ film and a flat InP(111)B substrate. Small vertical arrows mark positions of twin boundaries, formed perpendicular to the substrate; horizontal arrows and kinks in zig-zag lines mark positions of twin boundaries, formed parallel to the substrate.

Fig. 2: Cross-sectional HAADF-STEM image of an interface region of a Bi$_2$Se$_3$ film grown on a rough InP(111)B substrate with a simulated image inserted. A difference in contrast between the experimental HAADF-STEM image and the simulated image is present since roughness has been not included in the simulation.