Electronic Supplementary Information

Spontaneous formation of monocrystalline nanostripes in the molecular beam epitaxy of antimony triselenide

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A. Impact of Sb/Se flux ratio on the external dimesions of nanostripes

Several Sb_2Se_3 containing samples are grown on (111)B-GaAs substrates by following the growth procedure described in the main text with different Sb/Se flux ratios ranging from 0.1 to 1.0 for 10 minutes. In the presented series of samples Sb-flux is kept constant at the beam equivalent pressure (BEP) of 1.7 x 10^{-7} mbar, whereas only the Se flux is varied by changing the temperature of Se effusion cell. It is found that the external dimensions of the nanostructures for Sb/Se of 0.5 and 0.1 are very similar. However, the nanostripe growth is significantly slower in the case of low Se flux conditions, Sb/Se = 1. As effect, the nanostripes are quite well separated from each other and their average diameter is significantly smaller compared to the other two samples. The common feature of the three samples presented in Figure A1 is that Sb_2Se_3 is deposited in form of highly anisotropic nanostripes, which are aligned along three directions.

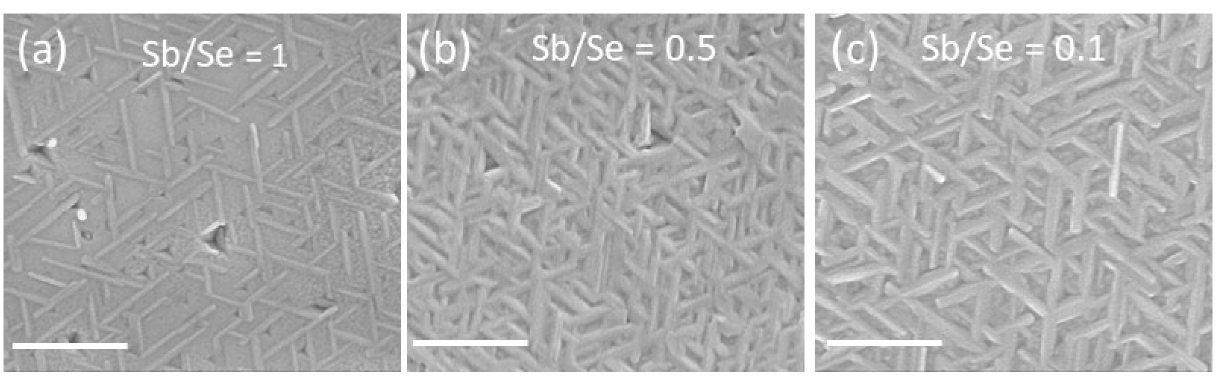

Figure A1 Scanning electron microscopy of antimony triselenide nanostripes grown on (111)B oriented GaAs substrates with the same Sb flux of 1.7 x 10^{-7} mbar and different Se fluxes of: (a) 1.7 x 10^{-7} mbar (b) 3.5 x 10^{-7} mbar (c) 1.6 x 10^{-6} mbar. All scale bars correspond to 200 nm. Data is collected with Zeiss Auriga scanning electron microscope operating at 5 keV with the probe current of 500 pA.

In order to obtain the information about the crystalline quality of these structures Raman scattering on the same samples is performed. For these measurements the spectra are excited at 532 nm at temperature of 12 K. The signal is detected using a CCD camera mounted on the output of a 0.75 m grating spectrometer (2400 grooves/mm). The spectra are presented in Figure A2. Spectral positions of the observed Raman active modes remain in agreement with the literature [29]. Parameters of the selected transitions in the Raman spectra are determined by fitting them with a sum of two Lorentz profiles. Obtained spectral widths are provided alongside the respective transitions in the Figure A2. The widths are found to be of the order of single cm^{-1}, which confirms a good structural quality of the NWs. They do not show a clear dependence Sb/Se flux ratio assisting the growth of the nanostructures.

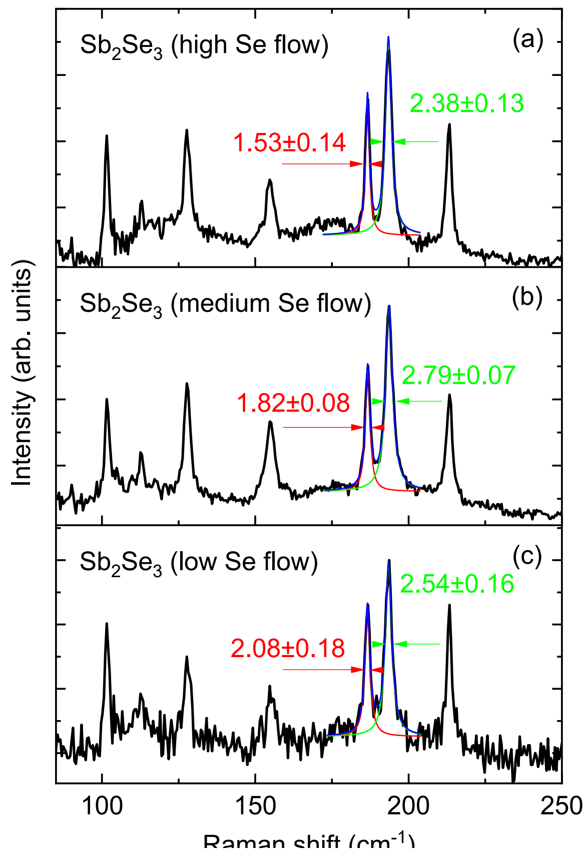

Figure A2 Raman scattering of antimony triselenide nanostripes grown on (111)B oriented GaAs substrates with the same Se flux of 1.7 x 10^{-7} mbar and different Se fluxes of: (a) 1.6 x 10^{-6} mbar (b) 3.5 x 10^{-7} mbar (c) 1.7 x 10^{-7} mbar. Excitation laser line is 532 nm at temperature of 12 K.

It is generally well established that the flux ratio in a mocular beam epitaxy process may significantly impact the crystalline quality and the crystal structure of the deposited layers. In the case of Sb_2Se_3, however, it is, most likely, the single phase crystalline structure that ensures a relatively small impact of this parameter on the growth process. At least our SEM and Raman scattering measuremets indicate this quite unusual effect.

B. Impact of substrate orietation on the external dimesions of nanostripes

For these investigations, three GaAs substrates with different orientations are mounted close to each other on the same holder. Sb_2Se_3 has been grown on them, simultaneously. The growth procedure itself is exactly the same as described in the manuscript. Sb flux corresponds to BEP of 1.7 x 10^{-7} mbar and Se flux – to 3.5 x 10^{-7} mbar. The growth temperature amounts to 320 ^{0}C and the growth time is 3.5 min. After the growth, the nanostripes are investigated by SEM. Their alignment is analogous to that presented in Figure 5. In this case, we focus, however, on the external dimensions of the nanostripes. A statistics of the observed Sb_2Se_3 nanostripes lengths and diameters for the three orientations of GaAs substrates are performed and are presented in Figure B1 on the left and right panel, respectively.

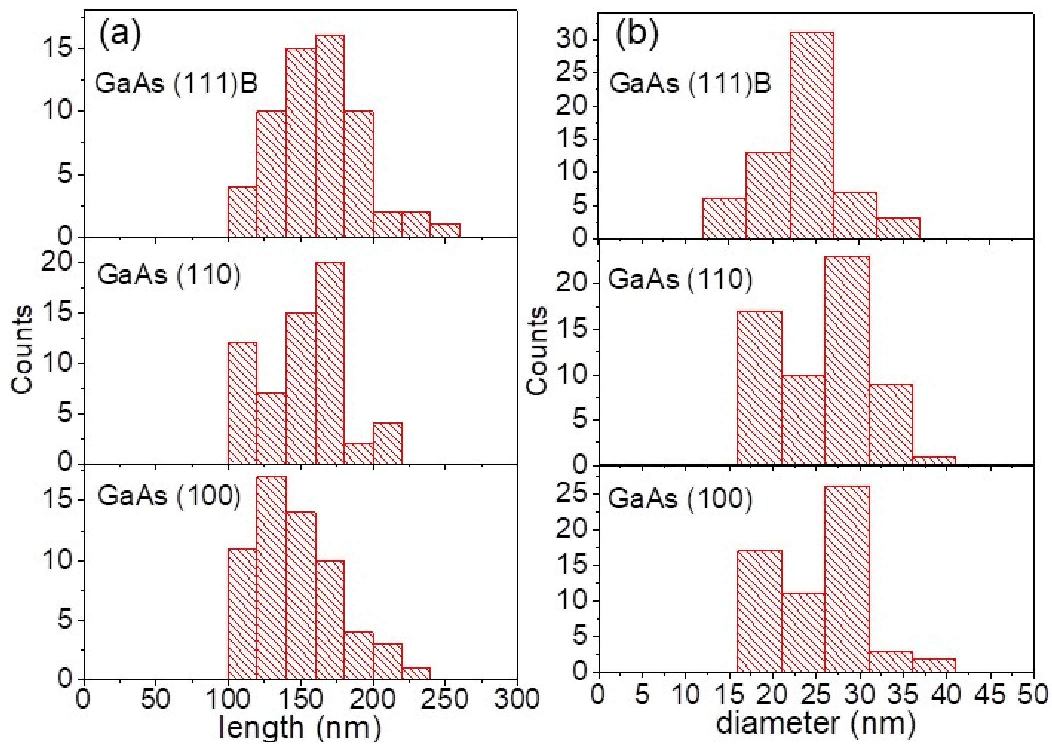

Figure B1 Statistics of nanostripe lengths (left panel) and diameters (right panel) of Sb_2Se_3 nanostripes grown on (111)B, (110) and (100) oriented GaAs substrates. The statistics is performed based on SEM images collected with Zeiss Auriga scanning electron microscope operating at 5 keV with the probe current of 500 pA.

The main conclusion from this experiment is that the sizes of the nanostripes do not change significantly when the growth is performed of differently oriented GaAs substrates.

The finesse of the Raman lines is known to reflect the structural quality of the structures. That is why Raman scattering has been performed on Sb_2Se_3 grown on differently oriented GaAs substrates. The results are presented in Figure B2.

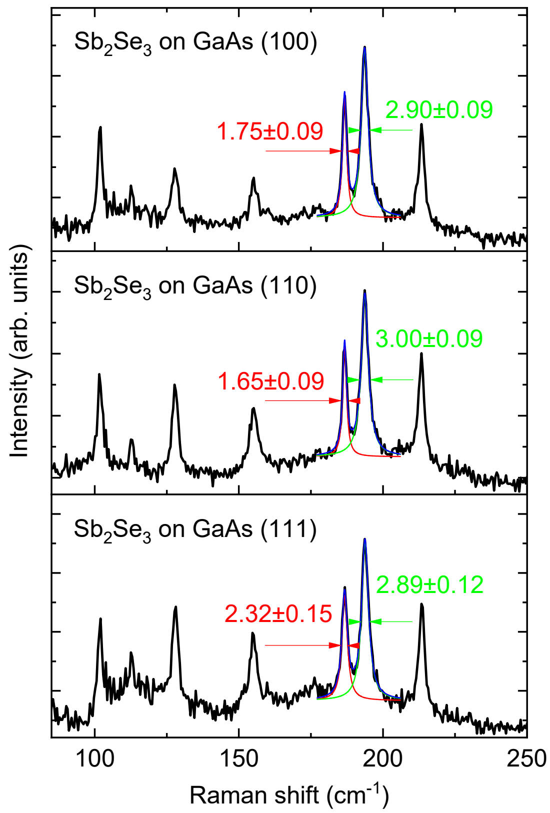

Figure B2 Raman scattering of Sb_2Se_3 structures grown simultaneously on differently oriented GaAs substrates. The finesse of the layers reflects the structural quality of this semiconductor. It is found that the finesse of the main line, marked in green does not change significantly depending on the crystal orientation of the GaAs substrate.

It is observed that the Raman spectrum, and in particular the finesse of the main line does not change significantly when Sb_2Se_3 is grown on differently oriented substrates. That is why we conclude that the Sb_2Se_3 structural quality is similar independently on the orientation of the substrate. What changes mostly is the nanostripes alignment, not their dimensions or crystalline quality.

C. Independent growth of different nanostripes

In order to verify whether the nanostripes are grown independently from each other or are formed on a continuous Sb_2Se_3 film a detailed analysis by HR TM has been performed. Figure C represents a typical HR TEM image with two different nanostripes. From these results it is concluded that there is not any continuous crystalline Sb_2Se_3 layer present between the nanostripes. This fact indicates that the nanostripes are not connected to each other and grow independently from each other.

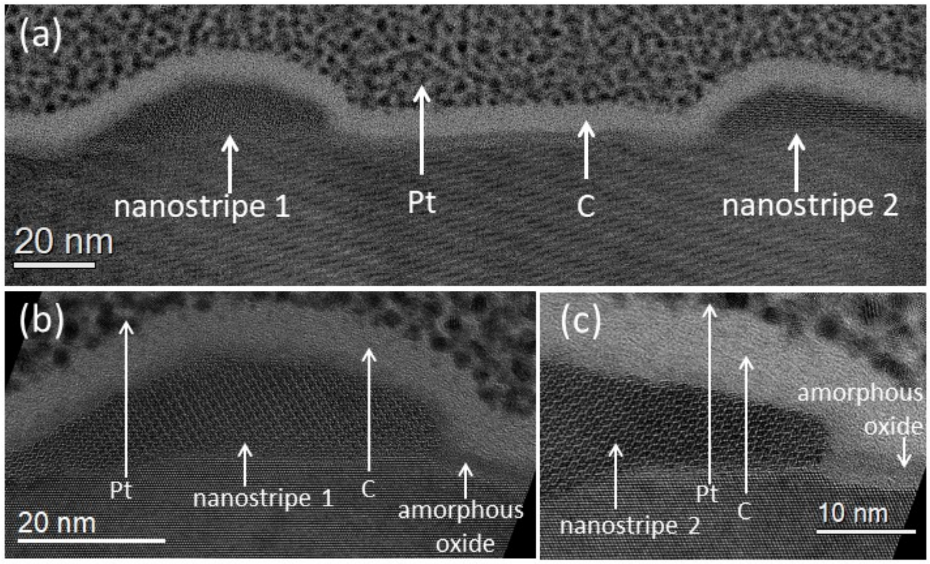

Figure C (a) Cross-section of two typical Sb_2Se_3 nanostripes measured by high resolution transmission electron microscopy.(b) and (c) Close-ups on the individual nanowires showing that there is no crystalline layer present between the nanowires. Platinum (Pt) and carbon (C) layers between the nanostripes come from the preparation procedure. Acceleration voltage is 300 keV.