Selective Area Growth of PbSe Nanostructures by Molecular Beam Epitaxy

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Selective area growth (SAG) by molecular beam epitaxy of PbSe offers exciting opportunities for integrated photonics and quantum technologies. By using an amorphous mask to define crystal growth, geometric control and deterministic placement of epitaxially smooth nanostructures can be achieved without etching, which can degrade quantum efficiency.[1] Integrating SAG with the advantageous properties of PbSe, including a narrow bandgap, high spin-orbit coupling, low Auger recombination rate, and a desirable defect tolerance,[3-4] could be instrumental for applications such as site-selective quantum dots, nanowire networks, and micro-light-emitting diodes in the mid-infrared. In this work, we explore PbSe SAG and characterize the promising growth morphology and optical quality.

Selective PbSe growth studies were performed over SiO_2 films patterned by e-beam lithography on Si-doped (001) GaAs with a flux supplied by a compound PbSe source equivalent to a growth rate of 0.42 Å/s. Preferred growth of PbSe in the mask openings was observed for substrate temperatures from 345-375°C with no polycrystalline growth on the mask at 375°C. Growth in 100×100nm^2 squares was observed to be largely dominated by one nuclei orientation, producing arrays of well-faceted and ordered squares with low-energy {001} sidewalls at ~365°C, even despite the large lattice mismatch. Atomic force microscopy confirmed smooth surfaces with an average 0.4 nm root-mean-square roughness within individual and 1.3 nm over all grains in Figure 1c. PbSe arrays showed room temperature photoluminescence in the mid-infrared, matching the planar emission wavelength. We aim to understand the structure of the nucleated islands with electron microscopy. This work was supported by the US Department of Energy (DE-SC0019274) and NSF (DMR-1906325).

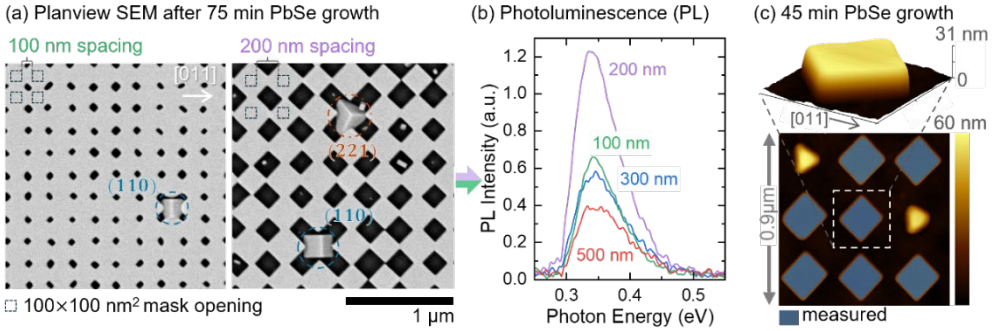

Figure 1. Planview scanning electron microscopy, (b) photoluminescence and (c) atomic force microscopy of PbSe growth in 100 nm square SiO_2 mask openings on (001) nGaAs.

[1] P. Aseev et al. Nano Lett. 2019, 19, 9102−9111 [2] J. Meyer et al. APL Mater. 9, 111112 (2021), [3] G. Springholz. Molecular Beam Epitaxy (Second Edition), Chapter 11, Elsevier, 2018. [4] P. Kumar, 8 - Efficient PbSe colloidal QDs for optoelectronics devices, Woodhead Publishing, 2022.

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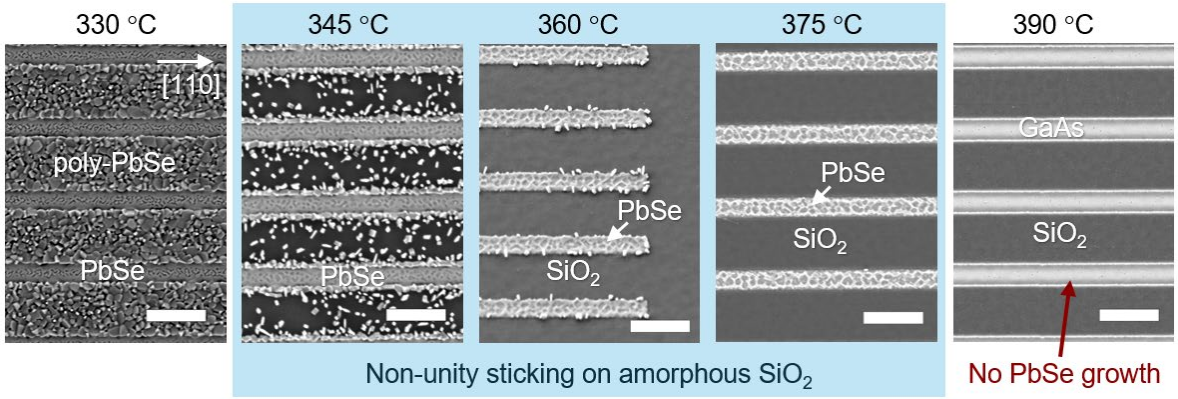

Figure 2. Planview scanning electron microscope (SEM) images after 20 min (~50.4 nm) PbSe deposition on 47 nm SiO_2 mask/nGaAs patterned by e-beam lithography at substrate temperatures varying from 330 °C, where complete coverage of the amorphous mask by polycrystalline material was observed, to 390 °C, where significant PbSe re-evaporation results in no growth in the GaAs window. Preferred growth of PbSe in the GaAs windows occurred between 345 °C and 375 °C. The scale bar is 1 μm.

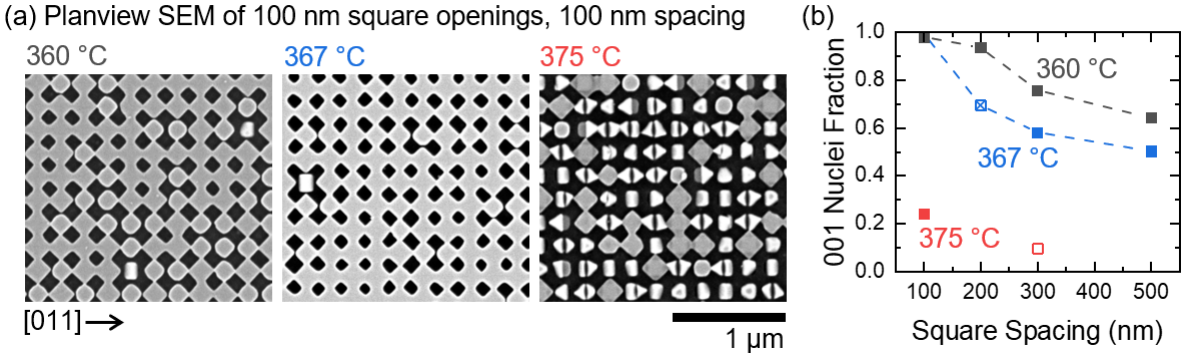

Figure 3. (a) Planview SEM of the produced nucleation growth morphology after 45 min (~113.4 nm) PbSe deposition on a 83 nm SiO_2 mask on nGaAs patterned with 100x100 nm^2 squares spaced by varying lengths. A decrease in cube-on-cube nucleation was observed with increasing substrate temperature as shown in the images and in (b) the graph of the fraction of (001) oriented nuclei versus the spacing between the GaAs windows for different temperatures. It is evident that the width of the mask may also play a role due to limited diffusion on the mask surface. (N_{sq} = population size, ■ N_{sq}>500, □ 200<N_{sq}<500, ⊠ N_{sq}<200)