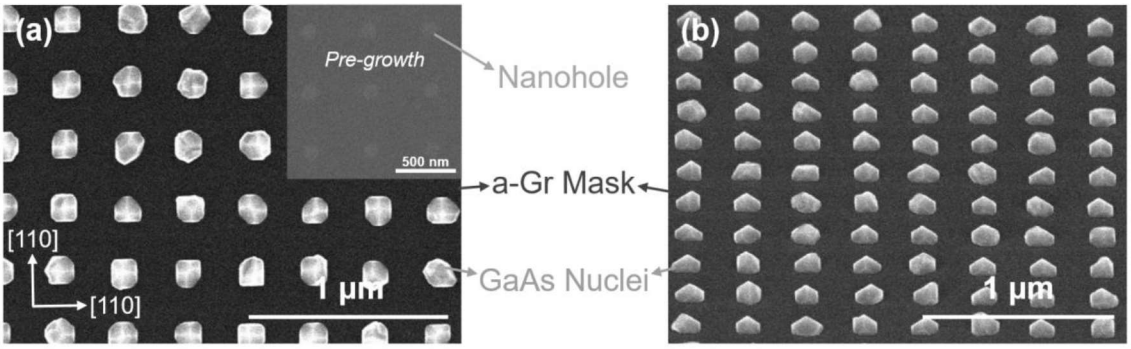


**Figure 1 (a, inset)** Nanoholes of ~100 nm diameter fabricated in a-Gr on Si. **(a)** Planview and **(b)** 45˚ tilted scanning electron microscopy (SEM) images from templated GaAs nuclei growth showing fully selective nucleation with complete hole filling. The faceting on the GaAs nuclei suggests high crystallinity.

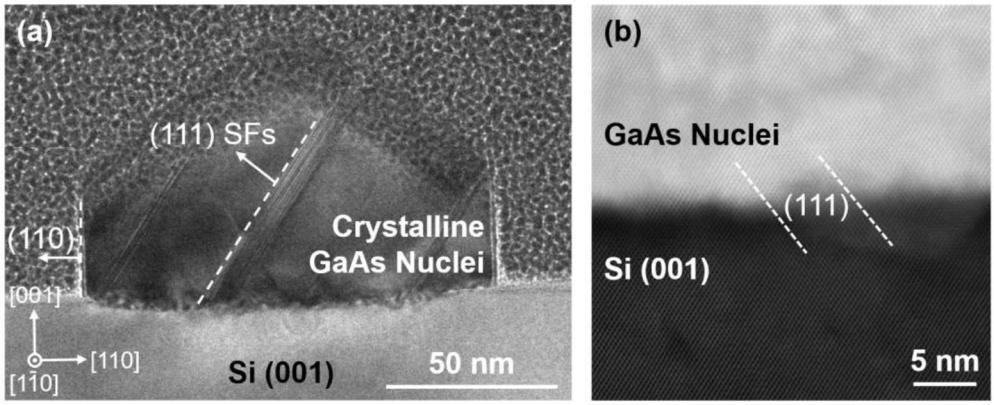


**Figure 2 (a)** High-resolution transmission electron microscopy (HR-TEM) images from a single-crystal GaAs nuclei with vertical (110) sidewalls. The nuclei exhibited stacking faults (SFs) and interfacial misfit dislocations. **(b)** High-angle annular dark-field scanning transmission electron microscopy image of the GaAs/Si interface of a crystalline GaAs nuclei showing clear registry of the GaAs nuclei to the Si substrate.

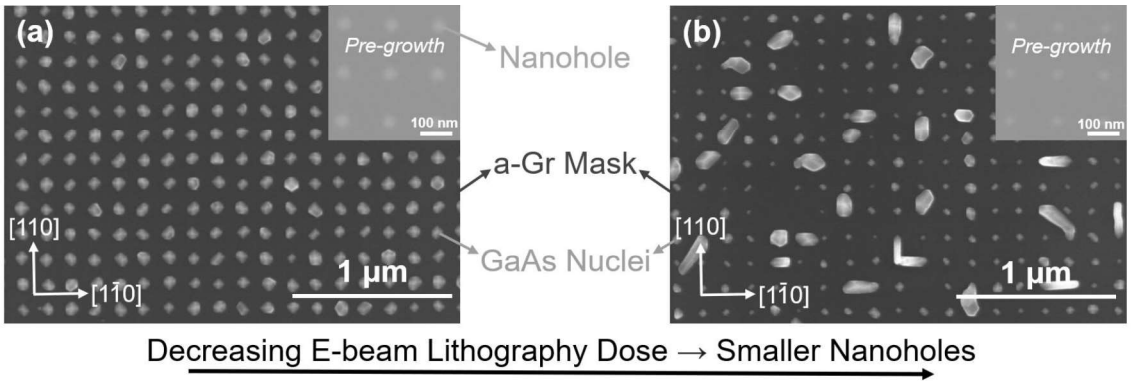


**Figure 3** Planview SEM images of GaAs nuclei grown in optimized arrays fabricated with varying e-beam lithography dose resulting in nanoholes ranging from **(a, inset)** ~35 nm in diameter for the highest dose to **(b, inset)** ~20 nm in diameter for the lowest dose. **(a)** Growth in nanoholes patterned under the highest dose exhibited selective, complete hole filling with uniform, faceted nuclei that were as small as ~50 nm in diameter. **(b)** Growth in nanoholes patterned under the lowest dose also exhibited selectivity with a high degree of hole filling while nuclei size fell into a bimodal distribution. The smallest nuclei were ~25 nm in diameter or less on the order of Stranski-Krastanov quantum dots. Larger nuclei exhibited early stages of lateral overgrowth beyond the edges of the nanoholes. These results underscore the delicate interplay between nanohole geometry and initial GaAs nucleation flux.