Selective epitaxy on nanoscale patterned areas is highly effective in the reduction of dislocation densities. Several theoretical studies have suggested that strain relief near the surface of the selectively grown epilayer results from three-dimensional relaxation mechanisms that are unavailable in large-area growth. These mechanisms for defect-free strain-relieved heterostructures become more effective as the epitaxial area is reduced. As a result of recent progress in strain-relieved heterostructures become more effective as the rel sheet 11011
diel position, the GaAs epilayer selectively deposited within each hole on the patterned substrate is surrounded by |1|011-type sidewalls perpendicular to Si(111), resulting in a hexagon-based prismatic pillar, without significant lateral overgrowth. At the initial stage of growth, twins parallel to Si(111) and an aperiodic mixture of cubic and hexagonal phases are observed but most of the GaAs pillars are terminated with a cubic phase region. Raman scattering reveals that the individual nanoscale GaAs pillars are completely strain relaxed. © 2005 American Institute of Physics. [DOI: 10.1063/1.1984100]

In this work, we investigate NPG of GaAs on Si(111) by molecular beam epitaxy (MBE). As seen below, a (111) plane is utilized for the fabrication of vertical GaAs nanowires with clear sidewall faceting. An advantage of a lithographic approach is the possibility of nanowires with uniform interwire spacings. Nanoscale periodic alignment of such wire-type epilayers has application to photonic crystals. Photonic crystals have been fabricated with etching techniques which suffer from scattering losses due to nonverticality and scattering from the etched surfaces. Monolithic integration of a periodic array of nontapered, strain-relieved, high-aspect-ratio GaAs nanowires on Si by epitaxial growth has a significant potential for Si-based photonics by providing active components with strict verticality and atomically smooth surfaces.

In this letter, we focus on the geometric crystal shape and strain relaxation at the initial growth stages of GaAs on a nanoscale SiO2-patterned Si(111) surface. Understanding of the initial growth mode and relaxation mechanisms is very important for future device applications.

The first step of sample preparation was MBE deposition of a ~20-nm-thick layer of amorphous (a) GaAs on an oxide-free Si(111) surface. Next a ~45-nm-thick blanket SiO2 film was deposited by electron-beam evaporation. A 355 nm period, two-dimensional (2D) circular hole pattern was formed on the SiO2 film with large-area i-line interferometric lithography and dry etching. For 300 nm deposition, the GaAs epilayer selectively deposited within each hole on the patterned substrate is surrounded by |1|011-type sidewalls perpendicular to Si(111), resulting in a hexagon-based prismatic pillar, without significant lateral overgrowth. At the initial stage of growth, twins parallel to Si(111) and an aperiodic mixture of cubic and hexagonal phases are observed but most of the GaAs pillars are terminated with a cubic phase region. Raman scattering reveals that the individual nanoscale GaAs pillars are completely strain relaxed. © 2005 American Institute of Physics. [DOI: 10.1063/1.1984100]

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and its dominance in faceting can be understood in the nanoscale regime.\(^6\) Thus, most of the GaAs nanopillars shown in Fig. 1(b) which show a hexagon shape have a prismatic shape, surrounded by six \{110\}-type sidewalls.

Figure 2(b) is a magnification of the region in the white box A in Fig. 2(a). As seen in this figure, a void is observed between the SiO\(_2\) mask and the Si substrate. A similar void is also observed on the other side of the nanopillar in Fig. 2(a). These spaces had been filled with the \(a\)-GaAs film, which evaporated from here as well as from the hole in the SiO\(_2\) during the 640 °C annealing process. While the voids of Fig. 2(b) are a limiting case and some of the \(a\)-GaAs film remains between the GaAs nanopillars to support the oxide mask, the formation of these voids confirms that the sacrificial \(a\)-GaAs film was removed from the circular holes before starting growth and does not affect the GaAs nanopillars.

In Fig. 2(b), lateral growth over the SiO\(_2\) mask is insignificant. The pillar shape of Fig. 2(a) implies that the growth rate along [111] is considerably greater than that along (110) on the SiO\(_2\)-patterned Si substrate. If growth is continued, a 2D array of high-aspect-ratio GaAs nanowires can be realized on Si[111]. This anisotropy of the growth rate is partly due to shadowing effects since the Ga flux is almost normally incident on the substrate, but more importantly is evidence that the equilibrium crystal shape for this nanoscale growth is a \{110\} bounded hexagon-based prism.\(^7\) Clear sidewall faceting with \{110\}-type orientations and high anisotropy of growth rate suggest the strong possibility of epitaxially grown, nontapered (e.g., strictly vertical), high-aspect-ratio, periodic geometric structures, which will be very important in the application to photonic crystals.

Figure 2(c) is a high-resolution XTEM image of the region inside the white box of the GaAs nanopillar shown in Fig. 2(b). In Fig. 2(c), a crystallographic twin observed at the very initial stages of growth is shown. This twin is generated by the 180° rotation of the crystal structure with respect to the growth direction (Si[111]). A twin is a typical crystal defect generated in a lattice-mismatched heterostructure and is known to contribute significantly to strain relief.\(^8\)

In Fig. 2(a), a few slightly contrasting stripes appear parallel to the Si substrate in the middle and lower part of the GaAs nanopillar. Figure 3(a) is a high-resolution image of the region within the white box B indicated in Fig. 2(a). A selected area electron diffraction (SAED) pattern taken near this region reveals two overlapped diffraction patterns—cubic and hexagonal phases. Comparison of Fig. 3(a) with Fig. 3(b) then implies that hexagonal phase is in the middle region of Fig. 3(a). In Fig. 3(b), the arrows indicate the spots from 6H–GaAs. According to this SAED pattern, the lattice parameter of 6H–GaAs along [0001], \(c\), is approximately 19.6 Å, exactly six times the (111) spacing of cubic phase, while the associated \(a\) is \(\sim\)4.0 Å. Then, the average spacing of these double GaAs layers of 6H–GaAs, corresponding to a lattice parameter of a typical hexagonal structure, is \(c/3\) \(\approx\)6.5 Å which is close to the theoretical result of 6.441 Å.\(^9\) In Fig. 2(a), the upper part of the GaAs nanopillar has a single

![FIG. 1. (a) A 45°-tilted and (b) a top view SEM image of the as-grown sample. The dashed circle in (b) corresponds to the original circular SiO\(_2\) opening. Once \{110\}-type sidewalls are fully developed, lateral growth slows and does not proceed significantly.](image1)

![FIG. 2. (a) A XTEM image of a single GaAs nanopillar. The arrows indicate hexagonal phase regions. (b) A magnification of the region inside white box A in (a). (c) A magnification of the region of the white box indicated in (b). The black arrow indicates a twin boundary.](image2)
The expansion coefficient is insignificant if the ratio of width to square-patterned GaAs/Si due to the mismatch of thermal prior to the appearance of the phase fluctuation. The formation of twins is observed in strain relaxation since the growth conditions, twins are presumed to play a major role in the GaAs nanopillars, respectively. The shift of the LO mode is exactly the same as that of bulk GaAs shown in the inset of Fig. 1, and implies that the GaAs nanopillars shown in Fig. 1 are almost fully relaxed.

According to theoretical modeling, the thermal stress in square-patterned GaAs/Si due to the mismatch of thermal expansion coefficient is insignificant if the ratio of width to thickness is comparable to or smaller than 1, as for the nanopillar shown in Fig. 2(a). This implies that misfit dislocations would not be formed after cooling from the growth temperature if they are not generated during growth. Freedom from thermal stress by geometric shaping, unavailable in large-area growth, is also another advantage of NPG and partly explains the absence of residual stress in the Raman results of Fig. 4.

In summary, we have reported NPG of GaAs on Si(111). On a 2D array of ~200–250-nm-diam circular holes opened with a 355 nm period on a SiO2-patterned Si(111) substrate, 300 nm selective deposition by MBE results in hexagon-based, prismatic GaAs nanopillars surrounded by six {110} sidewalls. At the initial stage of growth, twins and phase fluctuation between cubic and hexagonal phases are observed but most of the GaAs nanopillars are terminated with a cubic phase and are totally strain relaxed. These results have been interpreted as strain relaxation by phase fluctuation and twin boundaries and by a geometric shape that could be free from thermal stress in the nanoscale regime.

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