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Nucleation of single GaN nanorods with diameters smaller than 35 nm by molecular beam epitaxy

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Nucleation mechanism of catalyst-free GaN nanorod grown on Si(111) is investigated by the fabrication of uniform and narrow (<35 nm) nanorods without a pre-defined mask by molecular beam epitaxy. Direct evidences show that the nucleation of GaN nanorods stems from the sidewall of the underlying islands down to the Si(111) substrate, different from commonly reported ones on top of the island directly. Accordingly, the growth and density control of the nanorods is exploited by a “narrow-pass” approach that only narrow nanorod can be grown. The optimal size of surrounding non-nucleation area around single nanorod is estimated as 88 nm. © 2013 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4830044>]

Catalyst-free GaN nanorods/nanowires grown on Si substrates using molecular beam epitaxy (MBE) are reported as one of the very promising candidates for the next generation of nano-devices.^{1–3} The large window for growth-parameter variation, perfect crystal structures, and outstanding optical properties are the major attractions for the fabrication of III/V heterostructures monolithically integrated into the mature Si technology. For the reported growth of GaN nanorods, the nucleation density and aspect ratio of nanorods were controlled by the substrate temperature and III/V ratio,^{4–7} which also affect the crystal quality, the growth rate, and the morphology of rods. On the other hand, many groups have reported on the control of the rod density and position by the fabrication of a mask of a patterned oxide,⁸ SiNx,⁹ Si,¹⁰ or metal.^{11,12} The technologies of e-beam-lithography, UV lithography, or focused ion-beam technology are often used to selectively grow GaN nanorods.^{9,13,14} In these reports, multiple nuclei tend to appear at single defined site due to the large diameter of the mask openings. Spontaneously formed multiple nanorods were grown and successively merged into larger rods.^{9,10} The uniformity of the rod diameters decreases drastically for mask openings <300 nm due to the technical limitation of e-beam lithography and etching.⁹ Therefore, an understanding of the nucleation mechanism is crucial.

Many efforts have been carried out to investigate the nucleation mechanism of catalyst-free GaN nanorods/nanowires grown on Si or AlN surfaces by MBE. Some reports show that dislocations and plastic relaxation were necessary^{15–18} for the nucleation. However, reports based on the observations of the polarity of nanorods suggested that nanorods were formed as a continuation of underlying pedestals,^{8,12,19} as evidenced from the statistics of the rod-diameters in comparison with the underlying island-diameters.

Ga-polar,²⁰ N-polar,^{12,21,22} or the coexistence of both polarities^{19,23} of nanorods have all been reported. Direct evidence other than the rod-polarity is therefore needed to clarify the origin of the nucleation mechanism. The mechanism behind the rod elongation is generally agreed to be due to the diffusion induced mechanism^{10,24–26} and the conventional migration-enhanced epitaxy (MEE)²⁷ on the sidewall of the rod. The diffusion flux of the adatoms from the sidewall to the top is comparable to the deposition rate. A higher growth rate has been observed in narrower rod/wires compared to the wider ones. On the other hand, the nucleation mechanism and the factors determining the density of nanorods are complicated and is still ambiguous.^{15,18,24,25,28–31}

In this work, the samples were grown in a molecular beam epitaxy system (EpiQuest RC2100NR) equipped with conventional Knudsen effusion cells for Al and Ga, and RF-plasma nitrogen source (SVTA). Prior to the crystal growth, the (111) silicon was cleaned degassed at 900 °C, a 7 × 7 reflection high-energy electron diffraction (RHEED) pattern was observed, and a pure Si surface was obtained. An AlN layer was first grown at 800 °C under metal-rich conditions for 45 s with an Al/N ratio that was varied from 1.2 to 6.0 in order to control the wire density. The GaN was also grown at 800 °C under N-rich conditions with the plasma power at 110 W for 180 min. The structure was monitored *in situ* by means of RHEED analysis using KSA400 (k-Space Associates). Structural and compositional analysis was performed with scanning transmission electron microscopy and energy dispersive X-ray spectroscopy (STEM-EDX) in a double-corrected microscope (FEI Titan³ 60-300) operated at 300 kV. An electron transparent cross-sectional sample was prepared using a focused ion beam milling instrument (Carl Zeiss Crossbeam 1540 EsB). The substrate was kept stationary and not rotated during the whole process of growth. As shown in Figure 1(a), Al-rich AlN was grown before the growth of GaN. Al_xSi_{1-x} droplets and Al_xSi_{1-x}N islands were formed simultaneously on top of the Si(111) substrates at 800 °C.

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FIG. 1. Fabrication of the nanorods. (a) Al-rich AlN was deposited on the Si(111) substrate. (b) N-rich GaN was deposited. The Al-Si droplets were consumed by the nitrogen irradiation and incorporated into the Al(Si)N islands. The concave prints were left as the leftover of the consumed droplets. (c) The GaN nanorods and GaN compact layer are formed. The prints are represented as circles. The nanorods grew at the edge part of the underlying Al(Si)N islands inside the prints. (d) After 180 min of growth, the observed GaN nanorods grew to diameters of 25 nm (left) and 20 nm (right) with around 550 nm higher than the GaN compact layer.

It has been reported that at this temperature, Si is dissolved during the exposure of both Al (Ref. 12) and Ga.^{32,33} Eutectic is expected to be formed at this temperature. Si from the substrate is accordingly considered to be incorporated into the droplet during the growth of metal-rich AlN. After the growth of GaN in N-rich condition, prints of the droplets were formed at the site of droplets, as shown in Figures 1(b) and 1(c). The sizes of the drop prints and island inside are related to the sizes of the droplets. Simultaneously, GaN nanorods are grown on top of the islands. A rough and compact layer, denoted as the “GaN compact layer,” was formed outside of the prints with morphology of coalescent columnar structure as commonly reported^{1,2,12,19,25} in the literature. The side-view of a scanning electron microscopy (SEM) image (Figure 1(d)) represents the typical result after the growth. Both the nanorods and the compact layer grown along the *c*-axis were mainly aligned to a direction perpendicular to the substrate.

The prints and nanorods can be identified as black holes and small bright spots, respectively, in the SEM image of as grown sample, as shown in Figure 2(a). The 25° tilted SEM image (Figure 2(b)) shows that the small bright spots in Figure 2(a) are nanorods. All the nanorods are located inside the prints with different sizes. Statistic based on several SEM images taken at different positions of the sample was performed. Prints were found with zero up to three rods inside each print in Figure 2(c). 52% of the prints contain nanorods, of which 79% contain only one nanorod inside. According to the statistic, the optimal size of a print containing only one rod is around 88 nm. Almost 100% of the prints of that size were found with only one nanorod while the rate decreases with increasing print size. There are also several prints with a size randomly scattered from 200 nm up to around 900 nm with two or three rods inside. The largest print found without any nanorod, or GaN crystal inside is around 450 nm in size, which can be recognized as the diffusion length of Ga adatoms on the Si/SiN surface. This size is comparable with reported experimental observations of 400–500 nm,^{10,13} but much larger than the theoretically predicted value of 100 nm.³⁵

In order to further explore the mechanism behind the nucleation and growth, a large print area with multiple rods

FIG. 2. Relationships between the nanorods and the prints. (a) Top-view SEM image of the as grown GaN nanorods. Prints are visible as black holes in the image. Nanorods appeared as small bright spots are marked by white arrows. All nanorods are located inside the prints with various sizes. The scale bar represents 200 nm. (b) SEM image with 25° tilted angle of view. The nanorods pointed by the white arrows are mainly aligned perpendicular to the surface of the compact layer. (c) The size distribution of prints categorized by different numbers of nanorods inside the prints. 52% of the prints contain nanorods, of which 79% contain only one nanorod inside.

is selected for extended observation (Figure 3(a)). For prints larger than 450 nm, not only the probability for formation of multiple rods increased but also the probability to find large GaN crystal together with the rods. These crystals have the same height as the surrounding compact layer and therefore cannot be observed in the SEM cross-sectional image of Figure 1(d). Four cases with different kinds of nanostructures exist concurrently in this area (as pointed out by the arrows 1, 2, 3, and 4). Both the nanorods (spots 1 and 3) and the elongated nano-crystals (spots 2 and 4) show bright intensity at the edge of the underlying island/compact-layer structure, which in turn is darker caused by the height differences. After a thorough examination from all images of cross-sectional SEM, it is found that the heights of the elongated nano-crystals (spots 2 and 4) are with 45 nm to 90 nm, much less than the heights of the nanorods (spots 1 and 3) with 500 nm to 600 nm, in accordance with the diffusion-induced mechanism mentioned in the introduction. All bright nano-crystals are surrounded by black empty areas with sizes in the range from 50 to 150 nm. The widths of the elongated nano-crystals are 19 nm and 26 nm (for the spots 2 and 4), which are similar to the rod-diameters, 20 nm and 24 nm for the spots 1 and 3, respectively. The nano-crystals always grew along the edges of the underlying island. Therefore, the elongated length directions of the nano-crystals always match the edge of the underlying island (as can be seen for spots 2, 3, and 4). Perpendicularly, the width direction matches the radial direction of the underlying island. The SEM image shows that, in the radial direction, the widths of the elongated nano-crystals are independent of the sizes of the underlying islands. For spot 1, the underlying island is too small to be observed; however, the same growth scenario is expected in the same print in terms of atom diffusion and nucleation. Interestingly, for spots 2 and 3, the nanorods are located at opposite position in respect to the underlying islands, which exclude the possibility of shadowing effect to be the dominate factor of the nucleation. The positions of

FIG. 3. Relationships between the nanorods (nano-crystals) and the underlying islands. (a) SEM image of a large print which contains different kinds of growth cases as shown. The arrows 1, 2, 3, and 4 mark the locations of a nanorod on the small island, an elongated nano-crystal on larger island, nanorod on larger island, and an elongated nano-crystal on the edge of the compact layer, respectively. The widths of the nano-crystals (along the radial direction of underlying island) in cases 1–4 are denoted as 20 nm, 19 nm, 24 nm, 26 nm, respectively. On the other hand, the lengths of nano-crystals in the perpendicular direction of cases 1, 2, 3 are 20 nm, 60 nm, and 35 nm, respectively, with a much larger variation, and depend heavily on the size of the underlying islands. Notice that for case 2 and 3 the nanorods are located at different position comparing to the underlying islands. (b) STEM-EDX analysis was performed to investigate the spatial distribution of atoms, as shown by red (Ga), green (Al), and blue (Si) in the EDX map.

nano-crystals in respect to the underlying island are therefore considered as random since the size of the print shown in Figure 3(a) is smaller than the diffusion length of Ga atom on Si which is around 400 nm in this case.

Cross-sectional STEM-EDX was performed, with focus on the Al, Ga, and Si emissions, as shown in Figure 3(b). A nanorod-like thin layer of Ga signal was detected on the side-wall of the underlying island. Strong Ga signal was detected on top of the nanorod. The observations indicate that the nanorods shown in Figure 3(a) are not directly grown on the top of the island but originate from the sidewall of the underlying island all the way to the Si/SiN surface. Simultaneously, a Ga signal can also be detected at the region of the island top surface, but with a much weaker intensity, which is consistent with the observation from SEM that GaN also grow as a larger diameter columns on the top surface, but with a much lower growth rate than for the nanorods. No Ga has been found at the surface of the Si/SiN in the print.

The ability to control the wire density without a pre-defined mask is one of the major advantages of this approach. Here it can be achieved by only changing the III-V conditions during the growth of the underlying islands. A rise of Al/N ratio up to 6.0 will increase the rod density to $1.1 \times 10^9 \text{ cm}^{-2}$ (Figures 4(a) and 4(b)) due to the increase of the print density without affecting the alignments of the nanorods. In the typical case of GaN nanorods grown directly on top of the Si(111) substrate without any buffer layer, the diameters of the rods vary from around ten to several hundreds of nanometers. It has also been reported that the sizes of the underlying islands determine the diameters of the GaN nanorods grown on top of them and have to be controlled very precisely in order to achieve a high uniformity. However, based on the statistics of our sample (Figure 4(c)), the uniformity of the averaged rod diameter is much higher even after the increase of the rod density in consistency with the growth scenario described in Figure 4. The nanorod diameters always are found to be $< 35 \text{ nm}$ and the larger columns are found to have the same height as the surrounding compact layer, so they are either located at the edge of the prints and merge into the layer, or at the center of prints with the same (low) growth-rate/height as the

surrounding layer and cannot be observed in the SEM image from the side. Since all merged larger columns have similar heights very different from the heights of the nanorods, the utilization of conventional lithography becomes possible by coating/etching with the photoresist on it.

A growth scenario³⁴ as depicted from the above mentioned observations is illustrated in Figure 5. After the growth of the Al-Si droplet and the Al(Si)N island (Figure 5(a)) under Al-rich conditions, then GaN was grown under N-rich conditions (Figure 5(b)). The Si/SiN surface was continuously exposed to the Ga flux in the area during the growth, which facilitate the Ga adatoms diffusion on top of the surface to the island. A thin layer of GaN was expected to form both on the sidewall and the top surface of the island, as well as on the outside of the print. The difference in the chemical potential between the top surface and the sidewall of the nanorod³⁶ has been estimated to be 39 meV/atom,²⁶ which means that the adatoms diffused from the sidewall to the top of the island tend to be incorporated at the corner. It gives rise to a difference in the growth rate between the corner and other parts of the top surface and explains the often-observed phenomenon that the nanorods always are located at the corner of the underlying island. When the

FIG. 4. Density control of nanowires is demonstrated by varying the III-V ratio of the AlN layer. (a) The SEM images from bird's-eye view show the increased wire density of $1.1 \times 10^9 \text{ cm}^{-2}$. (b) The diameter distribution of nanorods obtained from statistics. The FWHM of the Gaussian fitted curve is 13 nm. (c) The density of the nanorods can be controlled by the variation of the Al/N ratio. An exponential growth function is used for the fitting.

FIG. 5. Illustration of the nucleation scenario. (a) The growth of the Al(Si)N island. (b) GaN was deposited under N-rich conditions. Ga atoms can diffuse from the Si surface to the sidewall and the top surface of the island. Concurrently, GaN crystals also form at the regular Si surface when the island is out of the diffusion range for the Ga atoms. (c) The GaN grows thicker with time. The nanorod was formed at the edge of the island by the elongation of GaN along one sidewall. (d) Upon continued growth, the nanorod will become taller due to a much higher growth rate (GR) compared to the region on the top surface of the island. The compact layer grew with a lower growth rate. The dimensions of 40 nm and 400 nm given in the figure can be varied by growth conditions.

growth continues, the difference gives rise to the elongation of nanorod, as illustrated in Figure 5(c). In the literature, the diffusion length on the sidewall (m-GaN) of the nanorod is reported to be around 40 nm (Refs. 25, 26, and 35) based on both experimental fits and calculation work, i.e., much smaller than the diffusion length of around 400 nm on top of the Si/SiN mentioned above. In the case of standalone GaN single nanorod directly grown on Si substrate without the consideration of underlying island, the nanorod growth rate has been modeled.³⁵ In the initial growth stage when the nanorod length L is much smaller than the effective diffusion lengths on the nanorod sidewalls λ_f (when $L \ll \lambda_f$), the growth rate of nanorod is independent upon λ_f and proportional to the square of the effective diffusion lengths on the substrate surface (λ_s^2), which induce a very high growth rate since λ_s is generally ten times larger than λ_f as abovementioned. When the nanorod grows much longer ($L \gg \lambda_f$), the growth rate of nanorod become independent upon λ_s and depends on λ_f ; therefore, a slower and constant growth rate is expected. In Figure 5(a), the whole growth process is depicted from $L \ll \lambda_f$, $L \sim \lambda_f$, to $L \gg \lambda_f$ in terms of the Ga contribution from the side wall of both the underlying island and the nanorod. In the initial part of the growth, it is important to keep the height of the underlying island smaller than the diffusion length on m-GaN (i.e., $\lambda_f = 40$ nm) to keep a high flux of diffusion adatoms from the Si/SiN surface to reach the upper corner of the island. The length can be varied by the growth condition. The higher the diffusion flux, the larger difference in growth rate between the position at the island corner and other parts of the top surface can be expected.

In conclusion, the origin of nucleation for the catalyst-free GaN nanorod on Si grown by MBE has been

investigated. Evidences show that the nanorods are not directly located on top of the underlying island as commonly reported, but stem from the side wall and the Si substrate at the bottom of it. A “narrow-pass” growth regime has been discovered and utilized to grow narrow (<35 nm), uniform, and density controlled nanorods, which is crucial for the advancement of nano-device fabrications.

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