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Nucleus and Spiral Growth of GaN Studied by Selective-Area Metalorganic Vapor Phase Epitaxy

Tetsuya Akasaka, Yasuyuki Kobayashi and Makoto Kasu

NTT Basic Research Laboratories, NTT Corporation,
3-1 Morinosato- Wakamiya, Atsugi, Kanagawa 243-0198, Japan
Phone: +81-46-240-3459 E-mail: akasaka@nttbrl.jp

1. Introduction

Atomically flat hetero-interfaces are crucial for developping high-performance semiconductor devices with single- or double-heterojunctions, or quantum wells. Recently, we have succeeded in fabricating step-free GaN surfaces with a diameter of 16 μm by selective area metalorganic vapor phase epitaxy (SA-MOVPE) [1]. The step-free GaN surfaces are atomically flat without any monolayer steps over the entire selective-area and therefore strongly expected to improve the performance of nitride-based quantum devices. Since the monolayer steps caused by the misorientation of the substrate are swept out from the selective-area growth, the growth mode (nucleus or spiral growth) is determined by the existence of screw-type dislocations for SA-MOVPE. This means that this technique enable us to evaluate the mechanisms of both the nucleus and spiral growth under the identical growth conditions on a single substrate. Here, we report the fabrication of step-free GaN hexagons with the diameter of 15, 30, and 50 μm. We also discuss the mechanisms of both the nucleus and spiral growth of GaN by SA-MOVPE.

2. Experimental

GaN films were grown by SA-MOVPE on GaN (0001) bulk substrates, which have selective-area masks with hexagonal-shaped openings with diameters of 15, 30, and 50 μm. The density of threading dislocations in the GaN template was less than $5x10^{6}$ cm^{-2}. Growth time and substrate temperature were fixed at 10 min. and approximately 950 ^{o}C, respectively. The source gases were trimethylgallium (TMG) and NH_3. The flow rates of TMG and NH_3 were $2.6x10^{-5}$ and $6.7x10^{-2}$ mol/min, respectively. The surfaces of GaN films were observed by atomic force microscopy (AFM).

3. Results and Discussion

Figure 1(a) show AFM images of a GaN film (hexagon) formed in a hexagonal opening of 50 μm in diameter. The GaN hexagon has a single wide terrace over almost the whole area. If a hexagonal mask opening has a screw-type dislocation, the obtained GaN hexagon exhibits completely different surface morphology [Fig. 1(b)]. This GaN hexagon has a large number of monolayer steps originating from a single screw-type dislocation. We observed a dislocation core at the left side of the hexagon and double growth spirals around the core as shown in Fig. 1(c). For the spiral shown in Fig. 1(c), we measured the interstep distance to be 0.135 μm. The degree of supersaturation, σ is derived from the interstep distance of growth spirals originating from a screw-type dislocation using a following equation,

$$\sigma = \frac{19W\gamma}{skT\lambda} \qquad (1)$$

where W is the volume of a Ga-N pair ($2.27x10^{-29}$ m^3), γ is the step energy (1.5 J/m), s is the number of growth spirals (2), k is the Boltzmann constant ($1.381x10^{-23}$ JK^{-1}), T is the growth temperature (1223 K), and λ is the interstep distance. The σ values are calculated for GaN hexagons by using equation (1) and plotted as a function of the diameter in Fig. 2. The values of σ are almost the same (~0.140) independent of the selective-area size used in this study.

The heights of GaN hexagons are plotted as a function of diameter in Fig. 3. We found that the heights are not changed by varying the diameter of hexagons both for the nucleus and spiral growth. However, the average height for the nucleus growth is 15 nm, which is approximately 30 times lower than that (430 nm) for the spiral growth. This means that the nucleus growth rate is strongly limited by the nucleation on step-free surfaces rather than by the lateral growth of nuclei. It can be seen in Fig. 3 that the nucleus growth rate is independent on the diameter of GaN hexagons. We therefore conclude that step-free GaN hexagons grow in the multi-nucleation mode. The growth rates by the multi-nucleation mode, R_n, are given by

$$R_n = aI_n^{\frac{1}{3}} v^{\frac{2}{3}} \qquad (2)$$

where a, I_n, and v are step height, nucleation rate, and the step velocity, respectively. The nucleation rates, I_n, of step-free GaN hexagons are estimated from R_n and v, where R_n is given by the heights of step-free GaN hexagons and v is measured from the height and the interstep distance of GaN hexagons grown in the spiral growth mode. I_n is plotted as a function of the diameter of GaN hexagon in Fig. 4. The obtained I_n is in a range of 10^5~10^7 $cm^{-2}s^{-1}$. The number of 2D nuclei generated during the formation of a GaN monolayer estimated from I_n is plotted as a function of the area of GaN hexagons in Fig. 5. From the slope of the linear fitting result (solid line), the 2D nucleus density is estimated to be ~$5x10^6$ cm^{-2}.

4. Conclusions

Step-free GaN hexagons with a diameter up to 50 μm were fabricated within a selective-area free of screw-type dislocations. The degree of supersaturation near the growing surface, σ, calculated from the interstep distance of the spirals was approximately 0.140, independent of the diameter of GaN hexagons. The nucleus growth rate of GaN was approximately 30 times lower than the spiral growth rate. The nucleus growth had a nucleation rate in a range of 10^4-10^6 $cm^{-2}s^{-1}$ and average 2D nucleus density of $5x10^6$ cm^{-2}.

Acknowledgements

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Reference

[1] T. Akasaka, Y. Kobayashi, and M. Kasu, Appl. Phys. Express **2** (2 009) 091002.

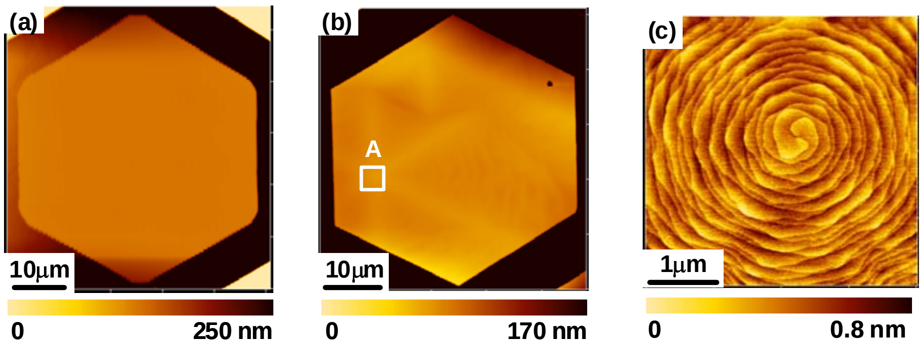

Fig. 1: AFM images of GaN films (hexagons) formed in hexagonal openings of 50 μm in diameter: (a) Step-free surface obtained within a mask opening without screw-type dislocations; (b) Surface with a growth spiral obtained within a mask opening with a screw-type dislocation; (c) Magnified scan image of the area in square A in Fig. (b).

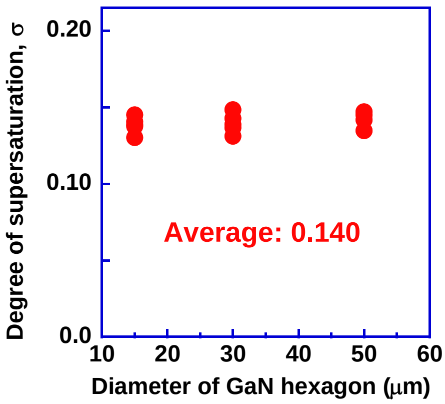

Fig. 2: Degree of supersaturation plotted as a function of the diameter of GaN hexagons, calculated from the interstep distances of growth spirals.

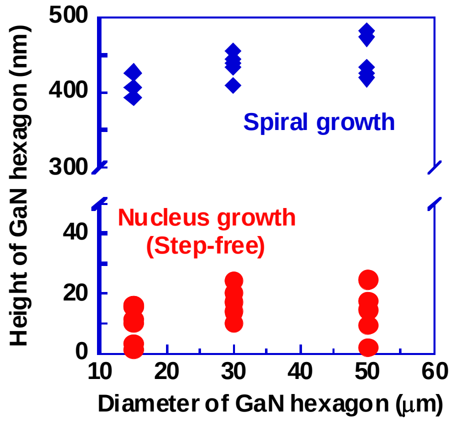

Fig. 3: Heights of GaN hexagons grown in the nucleus (closed circles) and spiral growth (closed diamonds) plotted as a function of the diameter of GaN hexagons.

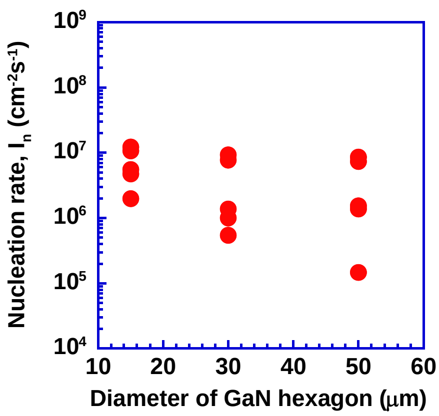

Fig. 4: Nucleation rate for the multi-nucleation growth of GaN plotted as a function of the diameter of GaN hexagons.

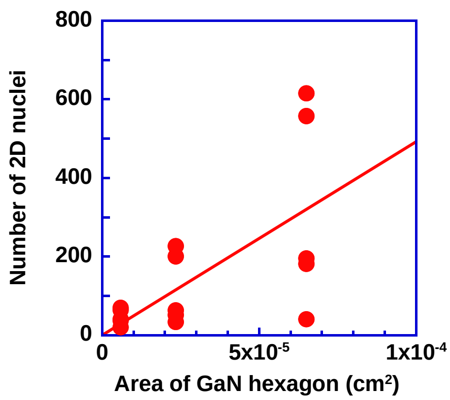

Fig. 5: Number of 2D nuclei generated during the formation of a GaN monolayer plotted as a function of the area of GaN hexagons.