

Surface Supersaturation in Nucleus and Spiral Growth of GaN in MOVPE

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1. Introduction

Atomically flat hetero-interfaces are crucial for developing 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 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, the growth mode (nucleus or spiral growth) is determined by the existence of screw-type dislocations. This means that this technique enables us to evaluate the mechanisms of both the nucleus and spiral growth under the identical growth conditions on a single substrate. Here, we report dependence of nucleus and spiral growth rates of GaN on the surface supersaturation in 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 16 μm . The density of threading dislocations in the GaN template was less than $5 \times 10^6 \text{ cm}^{-2}$. Growth time and substrate temperature were fixed at 10 min. and approximately 950 $^{\circ}\text{C}$, respectively. The flow rates of TMG were varied from 5.23 to 52.3 $\mu\text{mol/min}$, while the flow rate of NH_3 was fixed at $6.7 \times 10^{-2} \text{ mol/min}$. The surfaces of GaN films were observed by atomic force microscopy (AFM).

3. Results and discussion

GaN surfaces grown within selective areas having screw-type dislocations were formed by spiral growth and exhibited double growth spirals originating from dislocation cores, while those grown within selective areas without screw-type dislocations were formed by nucleus growth and had extremely wide atomic terraces. Figures 1(a)-(e) show AFM images of growth spirals obtained for GaN surface grown within selective areas having screw-type dislocations at various TMG flow rates. All the growth spirals are doubled and the step heights ($\sim 0.26 \text{ nm}$) correspond to the GaN monolayer. For the spirals shown in Fig. 1(a), the interstep distance was measured to be 0.303 μm . The interstep distances of growth spirals are plotted in Fig. 1(f). It can be seen that the interstep distance decreases

monotonically with increasing TMG flow rate.

Figures 2 show the growth rates of GaN hexagons grown in the spiral (a) and nucleus (b) growth modes, respectively. Both growth rates increase monotonically with increasing TMG flow rate. The growth rate for spiral growth, however, is much higher than that for nucleus growth. The result implies that the nucleus growth rate is strongly limited by nucleation on step-free surfaces rather than by the lateral growth of nuclei.

The surface supersaturation, σ , is generally derived from the interstep distance of growth spirals originating from a screw-type dislocation as follows [2]:

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

where W is the volume of a Ga-N pair ($2.27 \times 10^{-29} \text{ m}^3$), γ is the step energy (1.5 J/m^2), s is the number of growth spirals (2), k is the Boltzmann constant ($1.381 \times 10^{-23} \text{ JK}^{-1}$), T is the growth temperature (1223 K), and λ is the interstep distance. The σ values calculated using Eq. (1) increased monotonically from 0.0632 to 0.230 by increasing TMG flow rate from 5.23 to 52.3 $\mu\text{mol/min}$. The spiral and nucleus growth rates are plotted as a function of σ in Fig. 3. The spiral growth rate increases proportionally to σ^2 , while the nucleus growth rate is very low. Solid and dotted lines in Fig. 3 are fitting results we obtained using a crystal growth theory [3]. It can be seen that our experimental results are well explained by the crystal growth theory.

4. Conclusions

GaN surfaces grown within selective areas having screw-type dislocations were formed by spiral growth and exhibited double growth spirals originating from dislocation cores, while those grown within selective areas without screw-type dislocations were formed by nucleus growth and had extremely wide atomic terraces. The spiral growth rate increased proportionally to σ^2 , while the nucleus growth rate was much smaller than the spiral one.

Acknowledgements

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References

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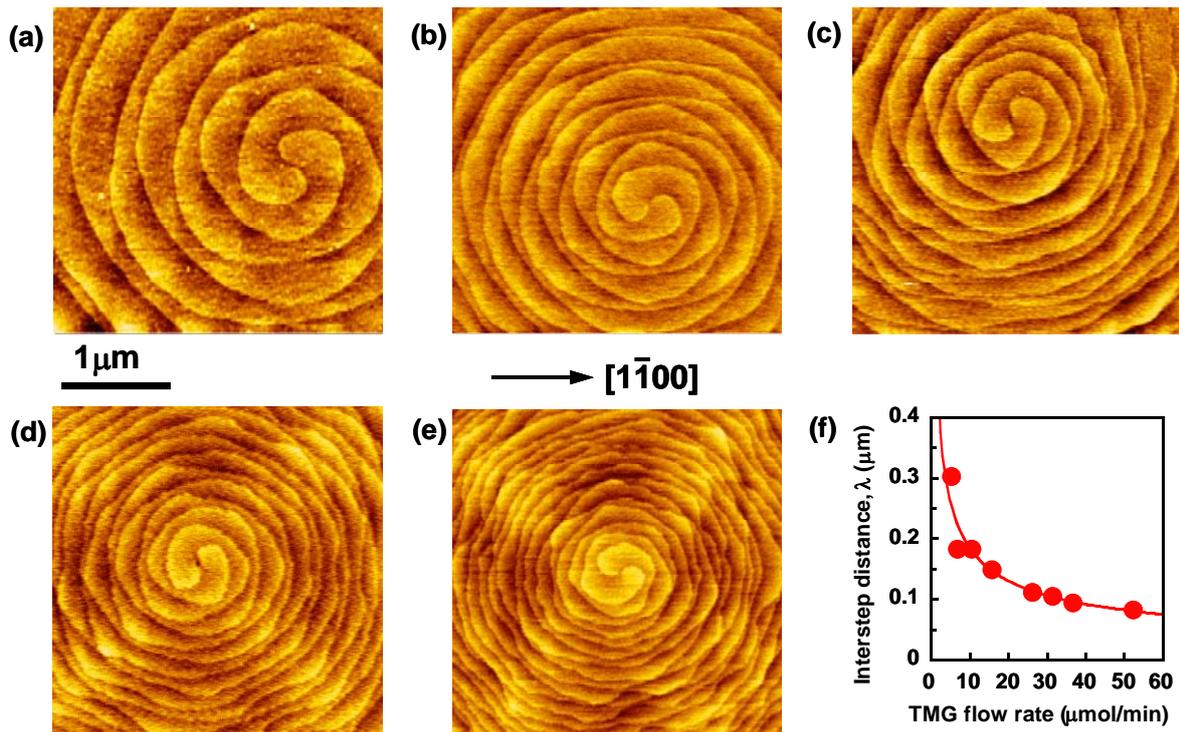


Fig.1: AFM images of growth spirals originating from screw-type dislocations: TMG flow rates of (a) 5.23; (b) 6.80; (c) 10.5; (d) 15.7; (e) 26.1 (μmol/min). (f) Interstep distance of growth spirals plotted as a function of TMG flow rate.

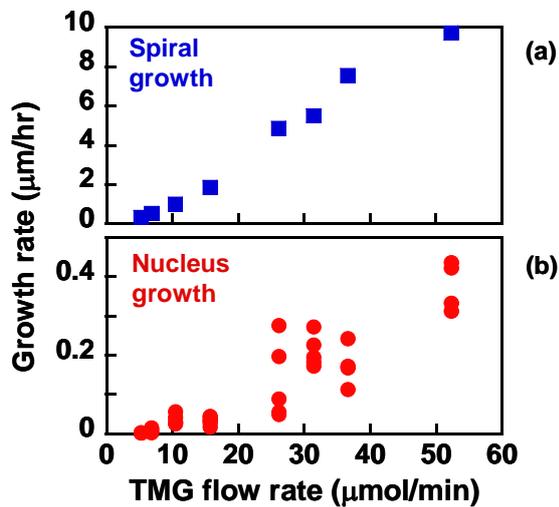


Fig. 2: Growth rates of GaN films plotted as a function of TMG flow rate. The films grew in: (a) spiral growth; (b) nucleus growth modes.

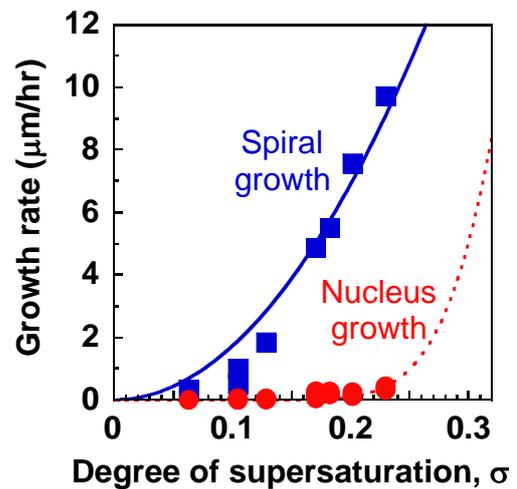


Fig. 3: Growth rates of GaN films plotted as a function of the degree of supersaturation for spiral growth (closed squares) and nucleus growth (closed circles). Solid and dotted lines are fitting results we obtained using a crystal growth theory