Large lateral growth rate in GaN grown directly on Al_2O_3(0001) substrate by two-flow metalorganic vapor phase epitaxy

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Developing two-flow metalorganic vapor phase epitaxy (TF-MOVPE), Nakamura et al. have grown high-quality nitride semiconductors and have succeeded in commercialization of blue light emitting diodes and laser diodes. [1)] However, they have not reported precisely how to grow high-quality films and why this technique is superior to conventional one-flow MOVPE. In order to obtain the information, we have investigated the direct growth of GaN on Al_2O_3(0001) substrate using TF-MOVPE. Once we establish the method to bring the large lateral growth rate, we will easily obtain high-quality films by inserting the buffer layer. [2)]

Figure 1 shows the schematic TF-MOVPE reactor used in this experiment. One main flow carries the reactant gas parallel to the substrate and the other subflow carries the inactive gas nearly perpendicular to the substrate. The characteristic of this reactor is as follows; The tilt of the subflow tube can be controlled precisely by the micrometers. Although the absolute values of the angle is difficult to determine, the relative angle can be determined within the accuracy of 0.01°. Here, we define the angle θ is a tilt from the vertical in the plane including this sheet and the main flow axis, and the angle ϕ is a tilt from the vertical in the plane perpendicular to this sheet. For the case of θ=1° and ϕ=0°, the H_2+N_2 gas flows toward the nozzle of the main flow.

The substrate was thermally cleaned at 1050 ℃ in a stream of hydrogen. Then, the substrate temperature was lowered to 1000 ℃ to grow GaN films. During the deposition, the flow rate of NH_3, trimethylgallium (TMG), and H_2 of the main flow were kept at 2 l/min, 38 μmol/min, and 1 l/min, respectively. The main flow speed was 45 cm/s. The subflow rate was 10 l/min, which corresponds to the subflow speed of 20 cm/s. Direct growth of GaN was carried out for 20 min at an atmospheric pressure.

The quality of GaN films depends on many factors such as the inclination of the main flow nozzle, main flow speed, V/III ratio, and subflow speed. In particular, it depends strongly on the angle between the axis of the subflow tube and the substrate. Figure 2 shows the interference micrographs of the surface of GaN films near the center of the substrate. Sample (a) is for the case of θ=1° and ϕ=0°. Normal hexagonal-like pyramid growth is observed. Although the roughness of the surface is ±0.5 μm, the average thickness is 2.6 μm. Sample (b) is for the case of θ=1.05° and ϕ=0°. Distorted hexagonal-like pyramid growth is observed. Many small pyramids indicate that in the sample (b) the lateral growth rate is smaller than in the sample (a). The average thickness is 2.0 μm. The tilt of 0.05° gives a large influence on both the lateral and vertical growth rates. Sample (c) is for the case of θ=1.30° and ϕ=0°. Three-dimensional island growth is observed. Surface coverage is 40% and the average thickness is 1.2 μm. For the case of θ=1.70° and ϕ=0°, no growth is observed in spite of the same flow rates.

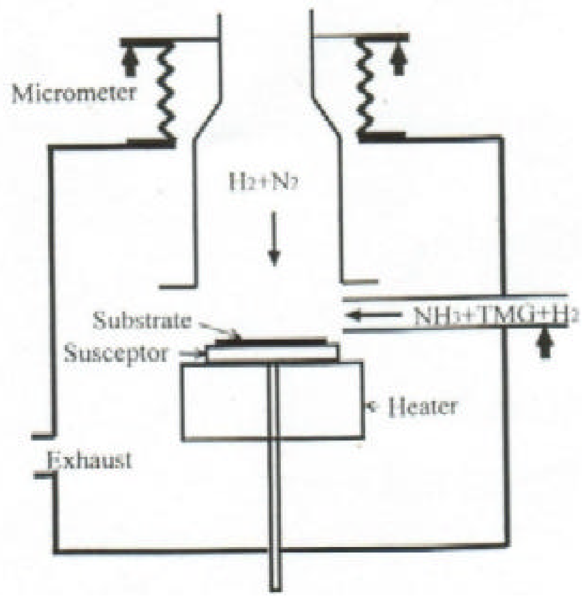

Fig. 1. Schematic TF-MOVPE reactor used in this experiment.

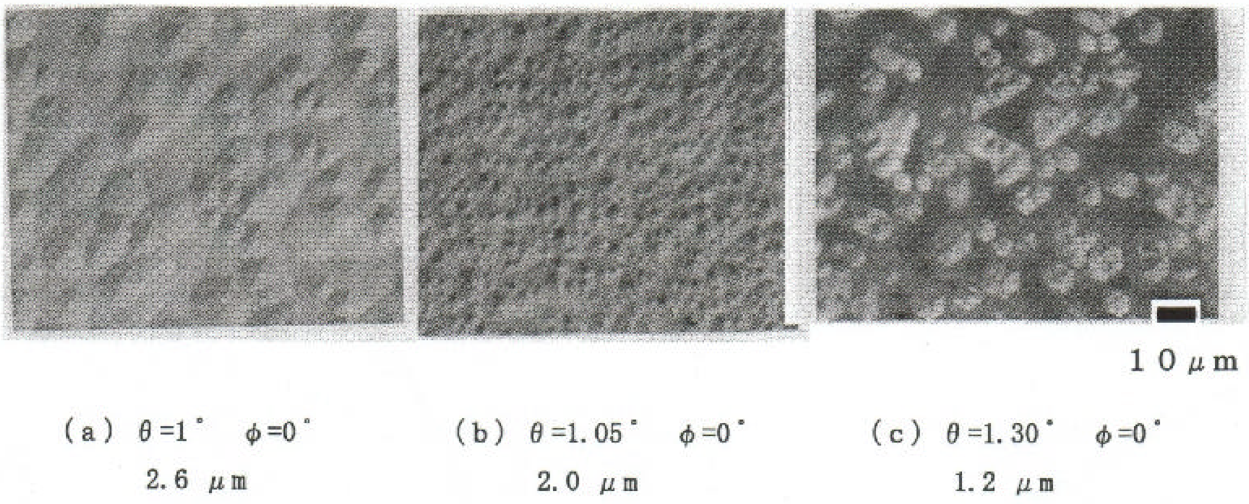

Fig. 2. Interference micrographs of GaN near the center of the substrate. The tilt of the subflow tube and the average thickness of GaN films are shown.

Near the center of the substrate, the similar results as in Fig. 2 are obtained regardless of the direction of the tilt. Near the edge of the substrate, however, the surface morphology depends on the direction of the tilt. For a case of $\phi \neq 0°$, the growth rate is larger near the narrower gap between the rim of the subflow tube and substrate. For the case of $\theta=1°$ and $\phi=0.05°$, for example, a continuous film is obtained at the narrower side, whereas three-dimensional island growth is observed at the wider side. For the case of $\theta \neq 1°$ and $\phi=0°$, no growth is observed in the downstream of the reactant gas. For the case of $\theta>1°$ and $\phi=0°$, three-dimensional island growth is observed at the upper stream.

The optimum angle depends on the main flow speed, the subflow speed, pressure, and so on. For the case of the main flow speed of 1m/s, for example, the optimum angle is $\theta=1.06°, \phi=0°$. The optimum angle θ increases with increasing the main flow speed.

The small tilt of the subflow tube gives a large influence on both the lateral and vertical growth rates of GaN films. When the subflow tube deflects from the optimum arrangement, GaN grows better at the narrower gap side, which indicates the reactant gas does not contact well with the substrate at the wider side. It is because the effect of the subflow is reduced and reactant gas drains faster from the wider gap. In order to promote the growth, it is important to bring the reactant gas into good contact with the substrate. Under the optimum arrangement, the reactant gas is considered to stay long on the hot zone, which leads to the efficient decomposition of NH_3, and to sweep on the substrate in all directions, which promotes both the lateral and vertical growth rates. It is the most important feature of TF-MOVPE that the reactant gas drains out on the substrate to all directions by adjusting the tilt of the subflow tube. Since the reactant gas flows parallel to the substrate, it drains out to all directions when $\theta=1°$ and $\phi=0°$. It is natural that the optimum angle depends on the main flow speed, subflow speed, pressure, and so on.

In summary, we have investigated the method to bring the large lateral growth rate in GaN grown directly on Al_2O_3(0001) substrate by TF-MOVPE. It is the most important to set the angle between the axis of the subflow tube and the substrate at a certain value ($\theta=1°, \phi=0°$) with the accuracy of 0.01°. Under the optimum arrangement, the reactant gas stays long on the substrate surface, which leads to the efficient decomposition of NH_3, and drains out on the substrate to all directions, which promotes both the lateral and vertical growth rate.

1) S. Nakamura, Y. Harada, and M. Senoh, Appl. Phys. Lett. **58**(1991)2021.
2) I. Akasaki, H. Amano, Y. Koide, K. Hiramatsu, and N. Sawaki, J. Cryst. Growth **98**(1989)209.