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Substrate Nitridation Effects on GaN Grown on GaAs Substrates by Molecular Beam Epitaxy Using RF-Radical Nitrogen Source

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The initial substrate nitridation effects on the crystal structure of GaN epitaxial layers grown on GaAs substrates by gas source molecular beam epitaxy using RF-radical N_2 as a nitrogen source was investigated. The crystal structure of GaN grown on (100) GaAs substrates was critically influenced by substrate nitridation, that is, the zinc-blende GaN was grown on the substrate without nitridation, and the wurtzite GaN was grown on fully nitrided substrate. These substrate nitridation effect observed here were contrary to the previously reported results using other nitrogen sources.

1. INTRODUCTION

III-V nitride semiconductors are one of the attractive materials for short-wavelength light emitting devices and highly stable devices under the severe environment (e.g. high temperature operation). Many efforts had been spent for clarifying the material properties and improving the crystal quality of nitrides[1]. Recently, it was known that the zinc-blende GaN could be grown on several cubic crystal substrates, such as (100) GaAs, Si, β-SiC, MgO, etc. This phenomenon attracted much attention from the points of view of material physics and engineering application. Fujieda et al.[2] have reported that the crystal structure of GaN grown by CVD using hydrazine was dependent on the initial surface nitridation of (100) GaAs substrates. Sufficient nitridation (longer than 600 sec) caused the zinc-blende GaN growth, and short time one (5 sec) produced the wurtzite GaN. Similar results were reported for GaN by GSMBE using dimethyl-hydrazine[3].

In this study, the GaN was grown by GSMBE using RF-radical nitrogen source on misoriented GaAs substrates from (100) toward [011] direction, with several substrate misorientation angles (SMAs). The crystal structure of GaN grown on (100) just GaAs substrates was investigated changing the initial substrate nitridation time t_N. The crystal quality was evaluated through the single crystal X-ray diffraction measurement, the surface morphology observation with Nomarsky microscope, and the room-temperature photoluminescence (PL) measurement. The crystal structure of GaN films was critically influenced by substrate nitridation degree, that is, the zinc-blende GaN were grown on the substrates without nitridation, and the wurtzite GaN were grown on fully nitrided substrates. These substrate nitridation effect observed here were contrary to the previously reported results obtained using other nitrogen sources[2],[3]. This opposite behavior in the substrate nitridation effect is reported here, for the first time.

2. CRYSTAL GROWTH

In the GSMBE system, the Ga beam flux was supplied from the conventional solid source, while the nitrogen beam was obtained through the RF(13.56MHz)-radical beam source using 100 % pure N_2 gas.

In the experiments, after the thermal cleaning followed by the growth of GaAs buffer layers at substrate temperature of 600-610° C, GaAs surfaces were exposed under As_2 and RF-radical N_2 for initial substrate nitridation. And then GaN was grown 90 minute at substrate temperature of 620° C.

3. RESULTS AND DISCUSSION

Figure 1 (a), (b), and (c) show X-ray diffraction patterns of GaN layers grown on (100) just GaAs substrates with the initial substrate nitridation time (t_N) of 0, 20, and 300 sec, respectively. In this case, the growth period (tg) and substrate temperature (T_{sub}) were set to be 90 min and 620° C, respectively. For the GaN without substrate nitridation (t_N=0 sec), the X-ray diffraction peak was observed at 40.1° which corresponded to the diffraction from the relaxed zinc-

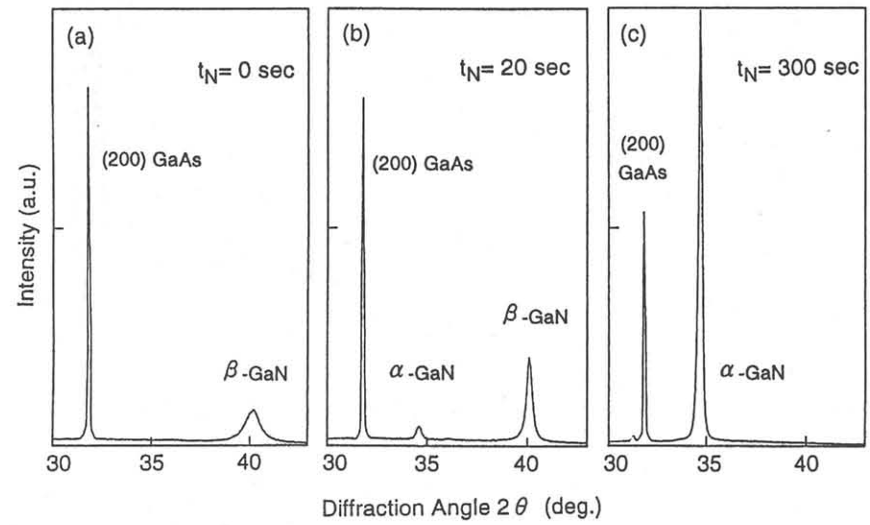

Fig.1. X-ray diffraction patterns of GaN grown on (100) just GaAs relation with different initial nitridation time (t_N). All samples were grown at 620° C, for 90 min, and the N_2 flow rate of 2.0 sccm.

blende (002) (β-)GaN (a=4.51 Å). On the other hand, for the sufficiently nitrided (t_N=300) case, the diffraction peak was appeared at 34.7°, corresponding to the (0002) diffraction from wurtzite (α-)GaN (c=5.18 Å). For the intermediate condition, i.e. t_N=20 sec, both peaks were observed suggesting that the wurtzite and zinc-blend structures were mixed. This phenomenon can be understood as follows. When there was no initial substrate nitridation, zinc-blende crystal structure of GaAs substrates can be directly transferred to GaN layer. On the contrary, in the long substrate nitridation case, the sufficientry nitrided GaAs substrate surfaces would be covered with stable wurtzite GaN, which may enhance the wurtzite GaN growth on that successively. While these results were opposite to the previously reported results given using different nitrogen sources. In those case, on fully nitrided substrates, the zinc-blende GaN was grown.

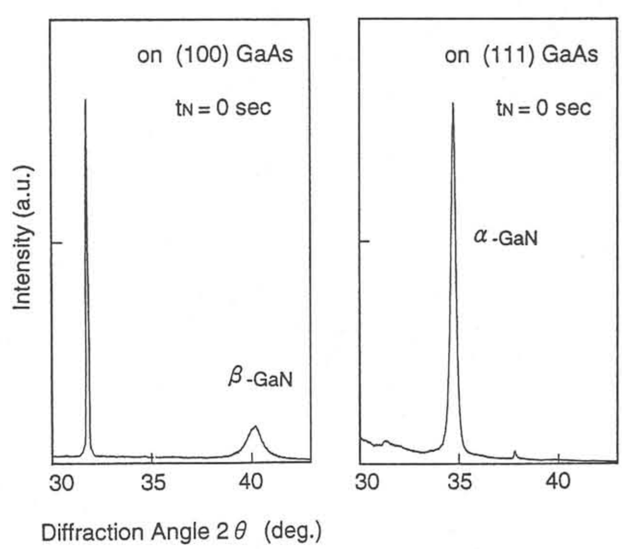

Fig.2. X-ray diffraction patterns of GaN grown on (100) just and (111)A GaAs without the substrate nitridation.

The narrowest full-width-at-half-maximums (FWHMs) of the zinc-blende and wurtzite GaN layers obtained in this work were 0.7° and 0.2°, respectively, which values were relatively narrow and Comparable with the best reported values[4].

On the other hand, it was confirmed that wurtzite GaN crystals were grown on (111)A GaAs without substrate nitridation. Figure 2 shows the X-ray diffraction patterns of GaN grown on (100) and (111)A substrates under the same growth conditions (t_N=0 sec), where the sharp diffraction peak at 34.7° (wurtzite) were observed for (111)A. Okumura et. al. reported the similar results, i.e. that wurtzite GaN was grown on (111)A GaAs substrates with the several minutes substrate nitridation, and under the same condition, on (100) GaAs substrate, zinc-blende GaN was grown[2].

The surface morphology of these GaN layers grown for 90 min are shown in Fig. 3. Defects were observed on the surface. As increasing the nitridation time, the defect size became larger (i.e. 0.5 μm in diameter at t_N=0 sec and 1.5 μm at t_N=300 sec), while

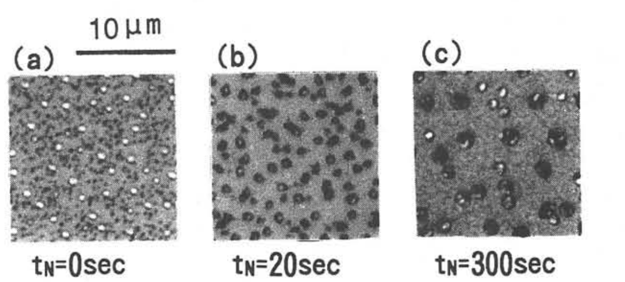

Fig.3. Surface morphology of GaN grown on (100) GaAs substrate with various initial substrate nitridation time. All samples were grown at 620° C, for 90 min, and N_2 flow rate of 2.0 sccm.

the defect density reduced. It was very difficult to measure the room temperature PL spectrum for these samples, because the high surface defects density induced the large surface scattering of the excitation light, and so the PL spectrum may be masked by the scattering light. The surface morphology of GaN layer was dependent upon the growth conditions, that is, the surface defect was reduced by increasing the growth time tg from 90 min to 180 min (compare Fig. 3 (a) and Fig. 4 (a)). Fig. 4 (b) shows the surface morphology of GaN grown on (111)A for 180 min. The defect density, in this case, was slightly reduced compared with (100) surface and the shape of defects were changed to triangle. While, for the samples with t_N=10-20 sec (condition for mixed wurtzite and zinc-blende structure growth), which were grown under higher V/III ratio, the surface morphology was drastically improved resulting in mirror like smooth surface, as shown in Fig. 4 (c).

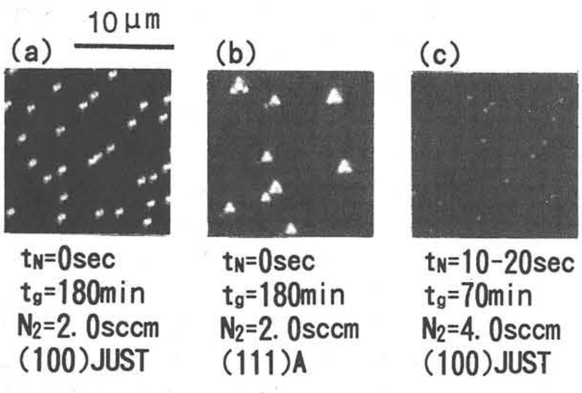

Fig.4. Surface morphology of GaN crystals.

To investigate the substrate misorientation effect, GaN layers were grown simultaneously on (100) just, 7° and 15° misoriented GaAs substrates from (100) toward [011] direction, and (111)A GaAs substrates, which all substrates were mounted on the same molybdenum block. The mirror like smooth surfaces like Fig. 4 (c) were obtained for all cases. The improved surface morphology of the GaN layers grown with higher V/III ratio enabled us to observe the PL peak at 365 nm. This peak corresponded to the bandgap wavelength of wurtzite GaN. Figure 5 shows the PL peak (at 365 nm) intensity as a function of substrate misorientation angle (SMA). Here the growth period was 70 min and the substrate temperature was 600° C. With increasing SMA, the peak intensity increased. This fact may suggest that the wurtzite crystal phase in epitaxial layers was increased and/or the crystal quality of wurtzite GaN was improved.

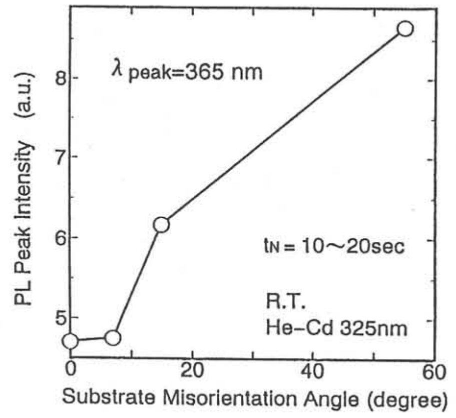

Fig.5. PL peak intensity as a function of substrate misorientation angle from (100) toward [011] direction. These GaN layers were grown at 600° C, for 70 min, and N_2 flow rate of 4.0 sccm.

4. CONCLUSION

In conclusion, the substrate nitridation effects on GaN crystal structure grown by GSMBE using RF-radical N_2 source was investigated. It was found that the growth without substrate surface nitridation allowed product of a zinc-blend GaN, which was contrary to the previously reported results using other nitrogen sources.

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REFERENCES

[1] see for example S. Strite and H. Morkoc, J. Vac. Sci. Technol. B, 10, (1992) 1237-1266.

[2] S. Fujieda and Y. Matsumoto, Jpn. J. Appl. Phys., 30, (1991) L1665-L1667.

[3] H. Okumura, S. Misawa, and S. Yoshida, Appl. Phys. Lett., 59, (1991) 1058-1060.

[4] K. Onabe, private communication.