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High quality GaN films - growth and properties

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Growth of GaN/Al_2O_3 layers by MOVPE has been investigated. Precise optimization of the growth parameters results in films with extremely high electron mobility: 900 cm^2/Vs at 300K and 4000 cm^2/Vs at 77K. The influence of the growth parameters on film properties like morphology, crystallographic structure, and the concentration of electrically active defects is presented. The mechanism of dislocation density reduction is proposed to explain the obtained results.

1 Introduction

Gallium nitride is a direct, wide bandgap semiconductor which has been intensively investigated over the last ten years, and has achieved practical success in optoelectronic devices like green/blue light emitting diodes and the blue laser [1] [2]. Despite its commercial success, many of the physical properties of GaN and its growth mechanism are still not well known. Usually GaN is grown on highly mismatched substrates like sapphire and silicon carbide using thin, low temperature, polycrystalline AlN or GaN buffer layers [3] [4]. A buffer layer gives dense nucleation on the substrate surface, but introduces strong disorder in the first hundred nanometers and a high density of dislocations in the final film [5]. The difference between the thermal expansion coefficient of the substrate and the film introduces stress during the cooling after the growth. This stress may causes additional dislocations.

Most papers dedicated to GaN heteroepitaxy describe the influence of the buffer layer on properties of the final film. The most important conclusion of these works is that the best film morphology can be obtained by using the thinnest possible buffer layer [4]. This approach is useful, but it does not explain the nature of the defects influencing the quality of the film. For example, the nature of electrically active defects supplying free electrons and defects influencing the electron mobility is still unknown. This paper presents relations between growth parameters and properties of obtained GaN films that provide information about these defects.

2 Experiment

GaN films were grown by the MOVPE method using a specially designed horizontal cell. To prevent parasitic reactions in the gas phase, reagents were mixed just before reaching the substrate. The growth was carried out on (0001) oriented sapphire substrate at atmospheric pressure using trimethylgallium (TMG), ammonia (NH_3) and hydrogen as the carrier gas. After annealing of the substrate thin GaN layer was grown at 500°C. Then, the temperature was increased to 1075°C for 15 minutes for recrystallization. The thickness of the buffer layer was experimentally optimized earlier to be as thin as possible to obtain good morphology of the final film. These buffer layer growth parameters were constant for all the experiments discussed here. The final layers of GaN were grown at a constant rate of about 2 µm/h in the temperature range 1020-1075°C. The III/V ratio was varied in the range 500-2500, keeping the total flow value constant (i.e. elevation of the ammonia flow was equalized by lowering of the hydrogen flow). Except as noted all films had a constant thickness of about 3 µm.

Electrical properties of the films were investigated by Hall effect measurement in the van der Pauw configuration in the range of temperature 30-400 K. Photoluminescence measurements were performed at 4.2 K using a He-Cd laser with output power 3 mW. Crystallographic properties were evaluated by X-ray diffraction for 00.2 reflection.

3 Results and discussion

The morphology of the films was mirror like except of those grown at the lowest temperature (less then 1030°C) or with the lowest V/III ratio (less than 750).

The surface of these films was partially perforated by pin-holes with hexagonal symmetry and diameter about 1µm. As it was shown by Hiramatsu *at al*. [5] the GaN layer growth starts from the three dimensional islands and through lateral growth forms the flat surface. Pin-holes can be the last trace of that process. We observed very high concentrations of pinholes on the layers with thickness below 3 µm, especially when grown at lower temperatures and low V/III ratio.

Electrical characterization of the investigated layers is shown in Figure 1. Electron concentration versus temperature shows an activation energy of about 14 meV. The electron mobility reaches a maximum close to 90-100 K. The dominant scattering mechanism at low temperature is connected with ionized impurities. The drop of mobility at very low temperature is due to a transition to hopping conductivity. It is well known that increasing the thickness of the films improves their quality. This improvement is usually explained as a reduction of the concentration of extended defects. For example increasing the thickness from 3 µm to 6 µm under the same growth conditions leads to an increase of mobility from 720 to 890 cm^2/Vsec at 300K and from 3100 to 3800 cm^2/Vsec at 77K.

The dependence of the electrical properties of the films on the growth temperature is shown in Figure 2(a,b). The electron concentration is almost constant, but low temperature mobility clearly increases with lower growth temperature.

The dependence of electrical properties on the V/III ratio is much clearer (Figure 2 (c,d)). Electron concentration increases with decreasing ammonia flow. This is surprising, because ammonia is potentially the most probable source of impurities. However, reduction of ammonia flow may cause a deficit of nitrogen in the layer. That deficit may lead via the creation of vacancies to an increased electron concentration. The change of mobility at 77K versus the V/III ratio does not correspond directly to the electron concentration. This suggests, that the mobility changes are not caused by the donor concentration only, but by other defects, for example dislocations.

Low temperature photoluminescence spectra (Figure 3) show absence of excitons bound to acceptors and very weak (more then 3 orders of magnitude less then exciton lines) donor-acceptor structure. This suggests a very low concentration of shallow acceptors.

Results of X-ray measurements are presented in Figure 4. It is seen that decrease of the ammonia flow down to a 750 V/III ratio decreases the mosaicity reflected in the rocking curve FWHM. Simultaneous increase of the strain is observed, which has to result from reduction of dislocations and grains boundaries. The growth when the V/III ratio is 500 corresponds to a film still perforated with pin-holes. Thus, the layer may have higher flexibility and it falls outside the trend discussed above.

The quality improvement observed for variation of the V/III ratio and growth temperature may be explained by the following mechanism. With a low growth temperature and low V/III ratio the rate of flattening of the surface is low. The pin-holes exists on the surface during the growth for a longer time and the probability of a dislocation crossing through the pin-hole face (wall) is higher. As was shown by Z.Liliental-Weber *at al*. [6], this crossing can cause bending and finally disappearance of dislocations. Apparently this reduction of dislocation density increases the electron mobility.

ACKNOWLEDGMENTS

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REFERENCES

[1] Shuji Nakamura, Takashi Mukai, Masayuki Senoh , *Appl. Phys. Lett.* ***64*** , 1687-1689 (1994).

[2] S Nakamura, M Senoh, S Nagahama, N Iwasa, T Yamada, T Matsushita, Y Sugimoto, H Kiyoku, *Appl. Phys. Lett.* ***70*** , 1417-1419 (1997).

[3] I. Akasaki, H. Amano, Y. Koide, K. Hiramatsu, N. Sawaki, *J. Cryst. Growth* ***98*** , 209 (1989).

[4] S. Nakamura, *Jpn. J. Appl. Phys.* ***30*** , L1705-L1707 (1991).

[5] K. Hiramatsu, S. Itoh, H. Amano, I. Akasaki, N. Kuwano, T. Shiraishi, K. Oki, *J. Cryst. Growth* ***115*** , 628 (1991).

[6] Z. Liliental-Weber, J. Washburn, K. Pakuła, J. Baranowski, “Convergent beam electron diffraction and transmission electron microscopy study of interfacial defects in gallium nitride homoepitaxial films”, Microsc. Microanal. 3 (1997)

FIGURES

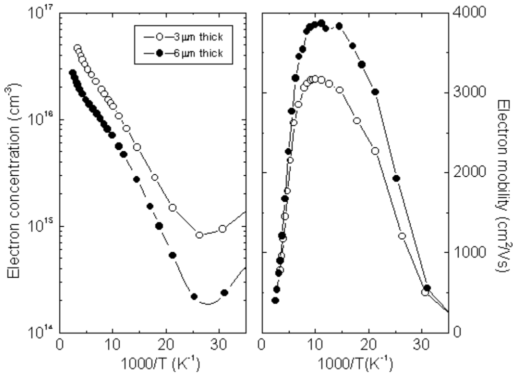

Figure 1. Hall electron concentration and mobility of two (3 and 6 µm thick) undoped GaN layers grown under the same conditions except for the growth time.

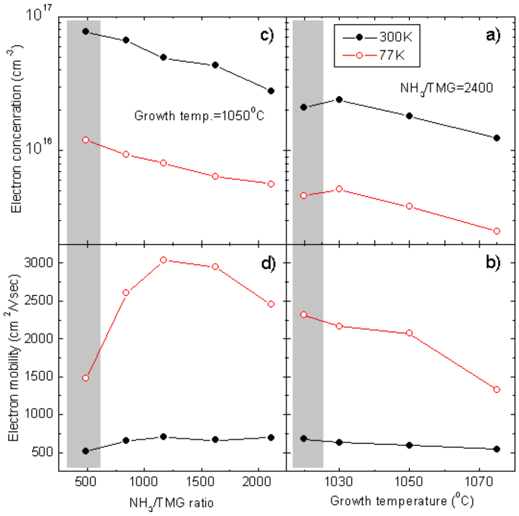

Figure 2. Electron concentration and mobility of 3µm thick layers as a function of growth temperature (a,b) and V/III ratio (c,d). The shaded regions indicate samples with pin-holes at the surface.

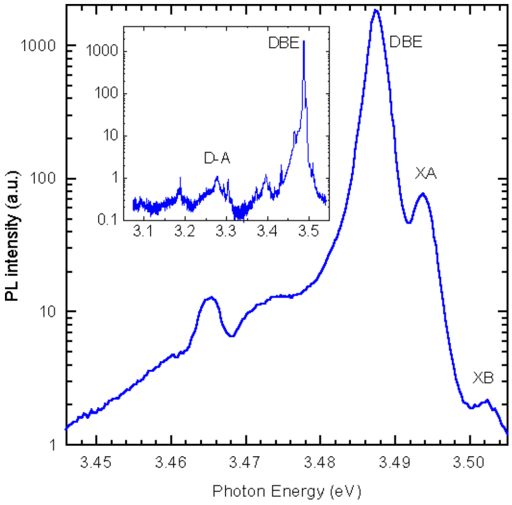

Figure 3. Low temperature (4.2K) photoluminescence spectra of the GaN layer. Free excitons (XA and XB) and donor bound exciton (DBE) are shown. Donor-acceptor (D-A) structure is seen in the inset.

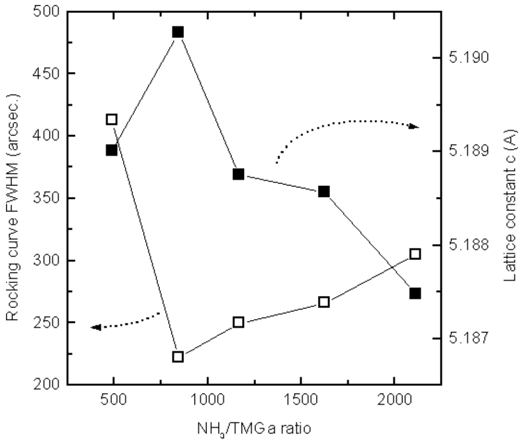

Figure 4. Rocking curve FWHM (left axis) and lattice constant c (right axis) of 3 µm thick layers as a function of the V/III ratio.