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Microstructure of GaN epitaxial films grown on (0001) sapphire by chemical beam epitaxy as related to buffer growth conditions

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Abstract. GaN buffer layers of about 20 nm thickness have been deposited on (0001) sapphire between 500 and 600 °C. The best morphology was found to correspond to a deposition made at 560 °C. Next, different annealing steps were made between 800 and 920 °C in ammonia or UHV. It is revealed that a partial desorption of GaN occurs when the buffer is heated up to 900 °C. In addition, three GaN epilayers of more than 1 μm thickness were grown on selected buffer layers after annealing. It is shown that the best results are obtained for a GaN epilayer grown on a buffer layer annealed at 900 °C in UHV, in agreement with AFM and optical measurements.

1. Introduction

Due to its wide direct band-gap, GaN is a promising material for numerous potential applications for optoelectronic and electronic devices. Although GaN and sapphire present a large mismatch in lattice constants (16.1%) and thermal expansion coefficients (−25.5%), high quality films on (0001) sapphire substrate are usually obtained using metalorganic chemical vapour deposition (MOCVD). Using this technique, several groups have succeeded in fabricating blue light-emitting diodes [1–3] and lasers [4]. The layers contain high densities of extended defects, such as threading dislocations and stacking faults. Although the major breakthrough was brought about by the discovery of p doping, the use of low temperature buffer layers played a key role as well. Since then, molecular beam epitaxy (MBE) has been used but with less success than MOCVD. Only recently has chemical beam epitaxy been introduced for the growth of GaN [5]. In the following, we report on structure of GaN buffer layers grown by CBE using triethylgallium (TEGa) and ammonia (NH_3) precursors, using various deposition temperatures and annealing conditions. Typical GaN epilayers grown on selected buffer layers were also analysed.

2. Experimental details

The GaN layers were grown on (0001) sapphire in a Riber 32 MBE system which has been modified for CBE. The gallium source was TEGa, with NH_3 as the nitrogen source. During the growth, the chamber pressure was $\sim 10^{-5}$ Torr. The sample description is given in table 1.

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Table 1. Growth conditions of buffer layers and epilayers.

Sample	Buffer thickness (GaN) (nm)	Deposition temperature (°C)	Annealing conditions
WB 36	20	520	
WB 31	20	560	
WB 45	20	600	
WB 66	20	560	NH_3/820 °C
WB 34	20	560	NH_3/920 °C
WB 33	20	560	NH_3/900 °C
WB 39	20	560	UHV/900 °C
	Thick GaN (1 μm) growth	Underlying buffer layer	
WB 22	820 °C	WB 66	
WB 52	850 °C	WB 33	
WB 42	850 °C	WB 39	

It should be noted that prior to the growth of buffer layers the sapphire surface was exposed to a short nitridation treatment at 900 °C [5].

Transmission electron microscopy cross-section samples were thinned down to 100 μm by mechanical grinding and dimpled down to 10 μm. Electron transparency was achieved by ion milling with a liquid-N_2 cold stage at 5 kV. A final step at 3 kV was used to decrease ion-beam damage. TEM and HRTEM observations were performed on a Topcon 002B electron microscope operating at 200 kV with a point-to-point resolution of 0.18 nm.

3. Results and discussion

3.1. Effect of growth temperature on the microstructure of buffer layers

First, a study of the morphological evolution of buffer layers with growth temperature is reported. GaN buffer layers were grown at 520 °C (WB 36), 560 °C (WB 31) and 600 °C (WB 45). It is clear from TEM analysis that the temperature has a significant effect on the surface morphology of buffer layers. All the samples are polycrystalline with mixed cubic/hexagonal phases. Grains with a pyramidal shape, characteristic of a three-dimensional growth mode, are observed in WB 36 and WB 45, and crystallites are found to be highly misoriented with respect to the sapphire substrate. In the case of WB 31 the layer is more continuous and rather flat (figure 1). It exhibits the minimum surface roughness for the series (the maximum is found for WB 45). These results are consistent with AFM observations [5], which indicate that the average grain size increases with the deposition temperature.

3.2. Effect of annealing conditions on buffer layers

Next, sample WB 31 was selected and annealed using various conditions. The aim was to obtain coalescence of the nucleation islands and therefore to have a continuous buffer layer made of very low angle grain boundaries [6, 7]. Two buffer layers grown at 560 °C were exposed to a short anneal of 3 min in NH_3 at 820 °C (WB 66) and 920 °C (WB 34). TEM and HRTEM observations of WB 66 revealed that the surface morphology changed: the large grains observed in the sample WB 31 became smoother. But large cubic areas crossing almost all the layer were present. On the other hand, for WB 34, no trace of GaN was visible in all the observed areas. The AFM study indicated that only a small fraction of the GaN had remained

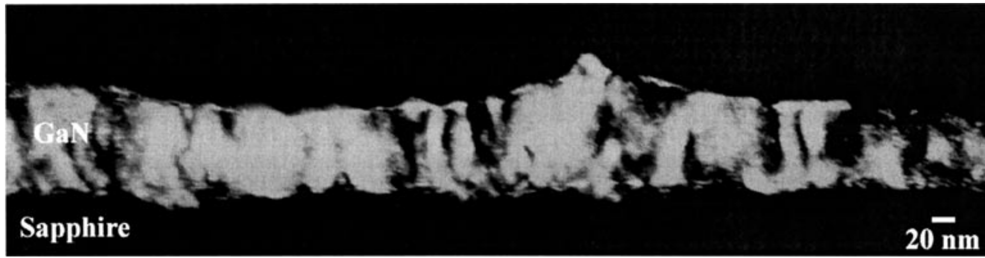
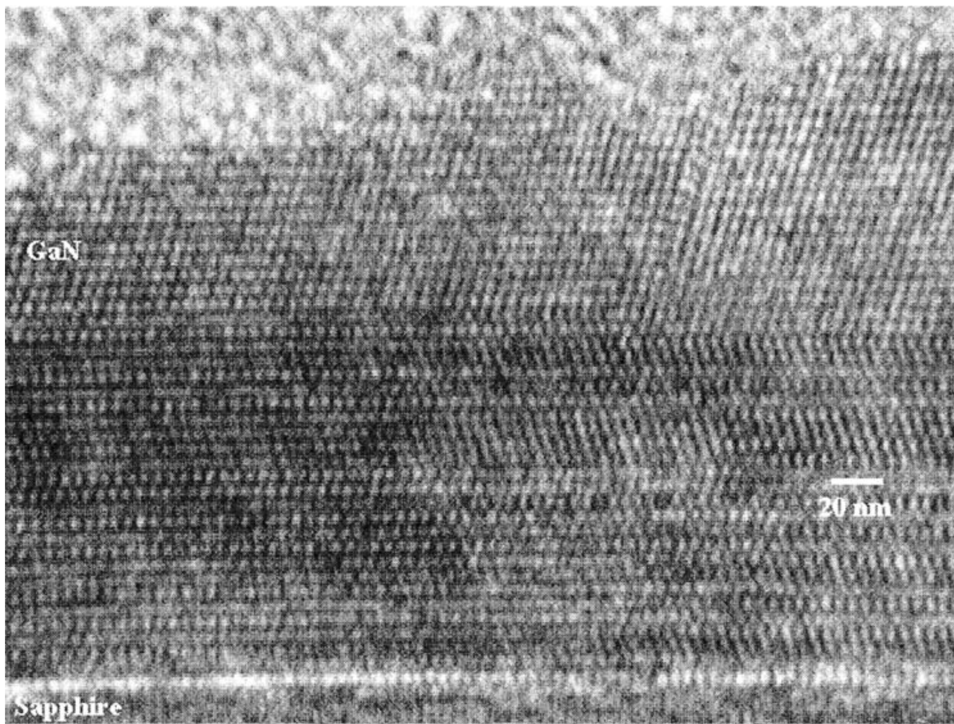


Figure 1. Dark field micrograph of the buffer layer grown at 560 °C (WB 31) showing a relatively flat and continuous surface.

(a)



(b)

Figure 2. (a) Bright field micrograph of WB 39. Partial desorption of GaN has occurred due to the annealing at 900 °C in UHV. (b) $[11\bar{2}0]$ HRTEM image of the buffer layer WB 39. Cubic phase is located at the top of the crystallites.

at the sapphire surface, meaning that the high temperature anneal has caused the GaN film to almost completely desorb. Two other buffer layers grown at 560 °C were exposed, this time to an anneal in NH_3 at 900 °C (WB 33) and in UHV at 900 °C (WB 39). For WB 33, the

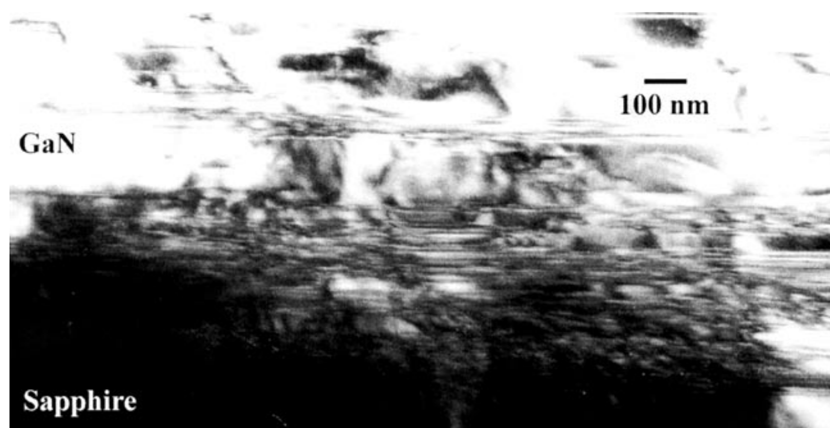


Figure 3. TEM image of the epilayer WB 52. A very high density of basal stacking faults cross the entire GaN layer.

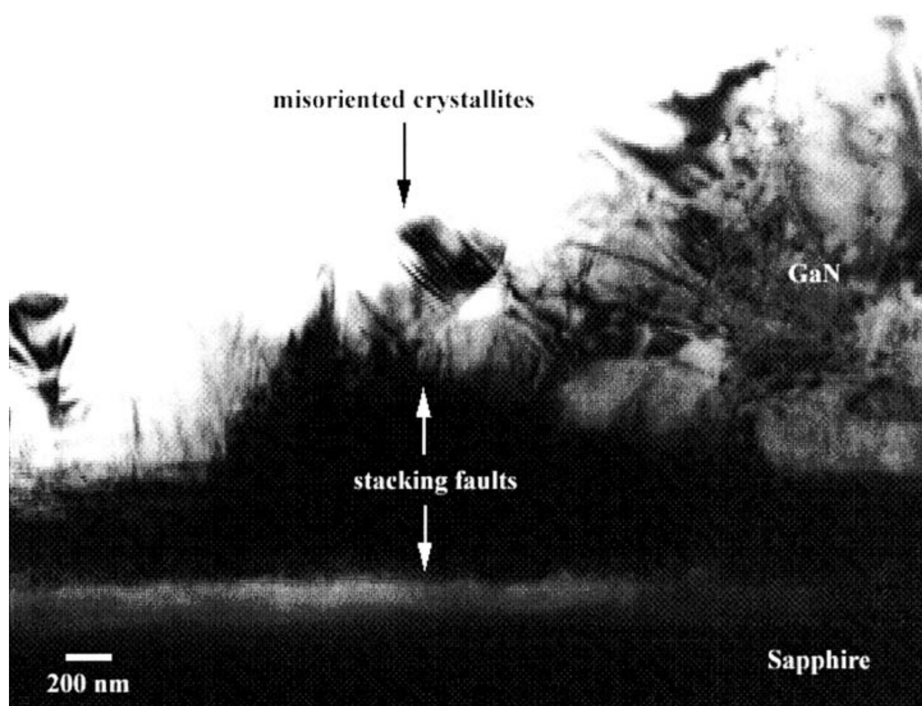


Figure 4. TEM image of WB 22 showing stacking faults and misoriented crystallites.

surface roughness of the layer appears to be important: although the structure is predominantly wurtzite type, there are nanometric amorphous areas. This loss of crystallinity may be due to surface contamination. This effect has already been observed by Vennegues *et al* [8], who suggested that oxygen incorporation may occur at low temperature by formation of Ga oxides that are subsequently desorbed during annealing.

No amorphous areas are observed when the sample is annealed in UHV at the same temperature (WB 39). The surface of this buffer layer is made of large islands, but it is

Figure 5. Weak beam image ($g = 0002$) of WB 42 showing the distribution of threading dislocations.

discontinuous (figure 2(a)). Once again, a desorption of the GaN film has occurred. The film is predominantly wurtzite structure and contains numerous stacking faults. Cubic parts are located at the top of each crystallite (figure 2(b)). Therefore, even in this case, the layer has not undergone a complete phase transformation from sphalerite to wurtzite.

3.3. The microstructure of the thick layers

Three epilayers of 1 μm thickness, WB 22, WB 52 and WB 42, were grown on top of the WB 66, WB 33 and WB 39 buffer layers respectively. The TEM analysis indicates that all the samples had a high density of basal stacking faults in the interfacial zone. For WB 52, a large area of these defects extends up to the top of the layer (figure 3), while for WB 22 the stacking faults are localized up to between 200 and 400 nm from the sapphire surface (figure 4), and only up to 50 nm for WB 42 (figure 5). It should be noted that for WB 22, the crystallites are largely tilted away from the growth direction.

Photoluminescence measurements have been carried out by Kappers *et al* [5]. The spectra taken at 10 K in the range of the band-edge showed a broad band at 3.47 eV (FWHM= 52 meV) for the sample WB 42, and a narrower band (FWHM= 20 meV) at the same energy for WB 52, accompanied by a number of smaller bands localized at 3.42, 3.39 and 3.29 eV attributed to excitons bound to dislocations. This emission band was not observed for the sample WB 22, but two other bands at 3.37 and 3.31 eV respectively were present and have been associated with extended defects in GaN [9].

Thus, optical properties of the epilayers are directly related to the structural quality of the buffer layer. The best results are obtained for WB 42, which presents the best crystalline quality and the lowest density of defects.

4. Conclusion

In this work, we have studied the microstructure of CBE GaN epitaxial films. It is shown that the structural and optical properties of these films are strongly correlate with the quality of

the buffer layers. This quality depends largely on the deposition temperature, but also on the annealing treatment. This annealing must be carefully controlled since GaN desorbs quickly at elevated temperatures.

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