Influence of buffer layers on the structural properties of molecular beam epitaxy grown GaN layers

V. Kirchner a,*, R. Ebel a, H. Heinke a, S. Einfeldt a, D. Hommel a, H. Selke b, P.L. Ryder b

a Institute of Solid State Physics, University of Bremen, P.O. Box 330 440, 28334, Bremen, Germany
b Institute of Materials Physics and Structural Research, University of Bremen, P.O. Box 330 440, 28334, Bremen, Germany

Abstract

The influence of low temperature buffer layers on the structural characteristics of GaN grown by molecular beam epitaxy on sapphire (0001) substrates was investigated. Layers grown on GaN and AlN buffers were studied by high-resolution X-ray diffraction and transmission electron microscopy (TEM). For both buffer materials, the variation of the buffer parameters, like their thickness and growth temperature, is reflected in a clear change of the GaN (0002) rocking curve width. For strongly decreased as well as for increased Bragg reflection width a deterioration of optical and electrical properties of GaN layers grown on buffers with respect to reference samples without buffer layers was observed. Moreover, layers grown on thin GaN buffer layers show extremely narrow ω scans and layer thickness interferences in 2θ/ω direction, while TEM reveals a high defect density throughout the entire layer. Therefore, not only the width of the rocking curves but also their shape has to be considered for the estimation of the defect densities by X-ray diffraction. © 1999 Elsevier Science S.A. All rights reserved.

Keywords: Buffer; GaN; Molecular beam epitaxy; X-ray diffraction

1. Introduction

The concept of low temperature (LT) buffer layer growth for the epitaxy of GaN on sapphire was developed in the eighties using metal–organic chemical vapour deposition (MOCVD) [1,2]. Today this approach is commonly accepted, because it allows the fabrication of GaN layers of good optical, electrical and structural quality. Both AlN and GaN are known to be suitable as buffer materials providing a high density of nucleation centers [3]. The influence of the various growth parameters like the thickness and the growth temperature of the buffer layer and the duration of the temperature-ramping process were investigated intensively [3,4]. Also, various models for the growth of the LT buffer were developed [5,6]. Whether these results and models are transferable to molecular beam epitaxy (MBE) with its significantly lower growth temperatures is not yet clear. Whereas some groups report an improved structural quality of MBE grown GaN on buffer layers [7–10], others omit the buffer without disadvantages [11,12]. Therefore, a study of the influence of AlN and GaN buffer layers on GaN layers grown by MBE is presented for selected sample series focusing on their structural properties using high-resolution X-ray diffraction (HRXRD) and transmission electron microscopy (TEM).

2. Experimental procedures

The growth was performed in a standard molecular beam epitaxy (MBE) system equipped with an EPI Unibulb radio frequency (rf) nitrogen plasma source. The plasma was operated at a discharge power of 500 W and a N2 flow of 1.5 sccm. The buffer layer growth proceeded in a temperature range between 380 and 785°C. In contrast to AlN buffers, GaN buffer layers were deposited on nitridated c-plane sapphire. The Al/N and Ga/N flux ratios were approximately 5/3 and 1, respectively, resulting in growth rates of 400 and 600 nm h⁻¹, respectively. The buffer thickness was varied between 3 and 150 nm and the thickness of the GaN layer was typically 660 nm.
X-ray measurements were performed with high-resolution diffractometers from Philips, equipped with a Cu sealed anode, a 4-fold Ge(220) monochromator and a 2-fold or 3-fold Ge(220) analyzer, respectively. For each sample, triple axis scans were measured in $\omega$ and in $2\theta/\omega$ or $\omega/2\theta$ mode for the GaN (0002) reflection. The scan direction in reciprocal space is the same for the last two modes but in the first case the intensity is plotted versus the $2\theta$ and in the second case versus the $\omega$ angle.

The cross-sectional specimen for TEM observation along $\langle 211 \rangle$ of the wurtzite GaN was prepared by mechanical grinding to about 5 µm thickness followed by ion milling. The TEM experiments were carried out on a Philips CM20/UT electron microscope operating at 200 kV.

3. Results and discussion

3.1. Growth without buffer layer

In Fig. 1 the (0002) $\omega$ and $\omega/2\theta$ triple axis scans of a GaN layer deposited without buffer layer (in the following referred to as reference sample) are shown. The used growth parameters were optimized within a wide range on the basis of the efficiency of the low temperature excitonic photoluminescence (PL) emission and the room temperature Hall mobility [13]. This optimization procedure resulted in typical values for the FWHM of the X-ray diffraction curves for about 1 µm thick layers of around 800 arcsec for $\omega$ scans and 80 or 40 arcsec for $2\theta/\omega$ or $\omega/2\theta$ triple axis scans, respectively. For the shown sample the exact values are indicated in the figure. It should be noted here that the (0002) peak width changed by a maximum factor of two for different growth parameters while the PL efficiency and the Hall mobility varied by orders of magnitude.

The strong difference in FWHM values in both scan directions is reflected in the typical elliptic shape of (0002) reciprocal lattice points, which has also been reported for MOCVD grown layers [14]. This shape is usually explained by mosaicity in the layers. A more detailed description of HRXRD results for layers grown without LT buffer layer can be found elsewhere [15].

3.2. Growth on AlN buffer layer

The FWHM of GaN (0002) $\omega$ triple axis scans for different growth temperatures of a 45 nm thick AlN buffer and different buffer layer thicknesses can be seen in Fig. 2. There is a clear reduction of the FWHM towards higher growth temperatures, but in comparison with the reference sample the values are larger for the whole sample series with varying buffer thickness. This behaviour contradicts the intended improvement by a buffer growth at low temperatures. The FWHM values also decrease with decreasing buffer thickness as shown in Fig. 2. In this case all buffers were grown at the temperature which provided the smallest FWHM in the preceding growth temperature.

![Fig. 1. GaN (0002) $\omega$ and $\omega/2\theta$ triple axis scans of a 1 µm thick GaN layer grown without a buffer layer.](image)

![Fig. 2. FWHM of the GaN (0002) $\omega$ triple axis scans in dependence on the growth parameters of AlN buffers. The squares correspond to layers on 45 nm thick buffers grown at different substrate temperatures, the circles correspond to buffers with varied thickness for the fixed substrate temperature of 785°C.](image)
series, i.e. 785°C. The minimum peak width of 480 arcsec for the 5 nm thick buffer is clearly below the value of the reference sample. However, the optical and electrical characteristics are inferior to those of the reference sample. All layers except those two on the thinnest AlN buffer layers were highly resistive, and their luminescence output was much weaker compared to the reference sample. The latter was true also for those layers which were characterized by (0002) Bragg peak widths clearly below the reference value [16]. This is a further hint, that the behaviour of the GaN (0002) peak width is not always in agreement with the optical and electrical data. Similar findings were already reported by other authors [17,18].

3.3. Growth on GaN buffer layer

Compared to the growth on AlN buffer, the use of GaN as buffer material leads to a much clearer decrease of the (0002) X-ray peak width in comparison to the reference sample. In Fig. 3 the FWHM values of the GaN (0002) ω triple axis scans for two sample series grown with different substrate temperature ramps between buffer and layer growth (see inset) are depicted in dependence on the buffer layer thickness. The FWHM obviously decreases towards thinner buffer layers for both sample series. Based on the model of mosaic layers, this indicates an increase in lateral coherence length or a reduction of the tilt of the mosaic blocks in the layer. The very small FWHM of 26 arcsec for the sample with a 3 nm thick buffer is comparable to extremely low values published by other groups [19,20]. Also, pronounced layer thickness interferences are observed in the 2θ/ω direction, as shown in Fig. 4, which have been described earlier [19,20]. The smaller the FWHM in ω direction the more pronounced the oscillations are. The interferences point to a large coherence length along the c-axis. A layer thickness of 630 nm can be determined from the distance of the interference maxima. As exemplarily shown in Fig. 4, all 2θ/ω scans of layers grown on LT GaN buffers of different thickness exhibit an asymmetry. Asymmetries of highly resolved diffraction profiles have been published several times [19,21], whereas different signs of the asymmetry were reported [19]. In our case, no asymmetry could be detected for samples grown without buffer (inset of Fig. 1), whereas GaN layers grown with an electron cyclotron plasma source as well as layers on AlN buffers show an increased intensity at the low angle side of the 2θ/ω scans in contrast to the behaviour in Fig. 4 [15]. The reasons for these effects are under investigation at present.

Cross-sectional TEM investigations of the sample with the 3 nm buffer layer showing the extremely narrow (0002) rocking curve reveal a very high defect density and a pronounced columnar structure over the whole layer as can be seen in Fig. 5. The defect density of this sample is at least one order of magnitude higher than that of the reference sample. A similar contradiction between defect density and diffraction peak width was found by Heying et al. [22]. In their case, this could be explained conclusively by the existence of pure edge...

Fig. 3. FWHM of the GaN (0002) ω triple axis scans in dependence on the GaN buffer layer thickness for two different growth temperature ramping regimes as indicated in the inset.

Fig. 4. GaN (0002) 2θ/ω triple axis scans for different GaN buffer layer thicknesses. The squares and circles in the inset correspond to the temperature regimes 1 and 2, respectively, shown in Fig. 3.
threading dislocations only which do not affect the symmetric reflections [22]. This was suggested by large FWHM of the (1012) reflection \( \omega \) scans as well as by TEM two beam images showing no contrast in \( g = (0002) \) case and strong strain contrast in \( g = (1120) \) case. In analogy to Heying et al. we also observed broad (1012) \( \omega \) scans but in contrast to their results a strong strain contrast was found in \( g = (0002) \) and \( g = (1120) \) TEM two beam images for our GaN layer on the 3 nm thick buffer, which is even more pronounced in the first case. To explain this contradiction, the rocking curves shown in Fig. 6 for three different buffer thicknesses might be helpful. For decreasing buffer thickness, these scans show more and more a remarkably different shape compared with the \( \omega \) scan of the reference sample in Fig. 2. The logarithmic plot in the inset reveals that this shape can be described by a superposition of a very sharp peak and a broad background. Probably, the latter is caused by the highly distorted parts of the crystal structure within the boundaries of the columns, each of which is nearly perfectly ordered in [0001] direction as indicated by the sharp peak. A detailed study of this problem is currently being performed.

The layers on thin GaN buffers are highly resistive so that their Hall mobility is not measurable, and the PL spectra are dominated by defect luminescence [16]. Considering the columnar structure of the investigated layers it is understandable that their luminescence efficiency and carrier mobility are much lower than those of layers grown without buffer.

![GaN layer](image)

**Fig. 5.** TEM bright-field micrograph taken along \( \langle 2110 \rangle \) GaN of a GaN layer grown on a 3 nm thick GaN buffer layer.

![GaN rocking curve](image)

**Fig. 6.** GaN (0002) \( \omega \) triple axis scans for different GaN buffer layer thicknesses. The squares and circles in the inset correspond to the temperature regimes 1 and 2, respectively, shown in Fig. 3. The rocking curve for sample A is additionally plotted on a logarithmic scale in the left inset.

4. Summary

The concept of LT buffer layer known from MOCVD growth of GaN on sapphire was applied to MBE. Both AlN and GaN were used as buffer materials. Changing the growth temperature and the thickness of the buffer layers strongly influences the structural properties of the GaN layers. This was reflected by GaN (0002) rocking curve widths varying by orders of magnitude. For layers grown on a thin GaN buffer layer extremely small FWHM values could be achieved. However, higher defect densities observed in TEM as well as deteriorated electrical and optical properties of the layers contradict the expected improved crystal perfection for layers grown on thin buffer layers. Consequently, the FWHM of the GaN (0002) \( \omega \) scan is not a proper indicator for low defect densities but the complete shape of the rocking curve has to be analyzed. Further intensive investigations are necessary to understand the presented data.

Acknowledgements

The authors would like to thank E. Golusda and S. Hesselmann for technical assistance. The work was supported by the Deutsche Forschungsgemeinschaft (Contract No. Ho 1388/10-1).
References
