Investigation of AlBGaN Structures for UV-Lighting

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In this work, a brief introduction in the advantages of AlBGaN as a material for UV-lighting devices will be given. As a template, AlN layers will be developed based on knowledge already obtained in our group. Also new approaches will be discussed to further increase the AlN crystal quality.

1. Introduction

Highly efficient blue light-emitting diodes (LED) already exist on the basis of Ga(In)N [1] enabling the generation of white light. Also LEDs in the UV range, especially the UV-C range (~ 270 nm), have many applications since they can be used for sterilization and disinfection of water and air [2, 3], phototherapy [2], flourescence analytical systems [3], bio-agent detection [4], spectrometry [5] and currency validation [5]. However, LEDs in the UV spectral range are typically much less efficient [2] due to the challenges of growing high quality AlN layers with low threading dislocation density (TDD). Such dislocations penetrating through the optically active layers, drastically decrease the efficiency of the device. Due to lattice mismatch between the sapphire substrate used for overgrowth and AlN, defects are generated. To minimize lattice mismatch between the substrate and the AlGaN with very high Al-content as well as reducing the absorption of light in the substrate, UV-LEDs are not grown on GaN- but on AlN substrates [6]. Moreover, lattice mismatch between layers of different Al content lead to further problems, e.g., further defect generation or even cracking. Due to the quantum confined Stark effect (QCSE), piezoelectric fields reduce the overlap of the electron and hole wave functions and, hence, the recombination probability in the active quantum wells decreases.

Boron containing AlGaN layers enable a further degree of freedom in lattice matching the optically active layers to the AlN templates since BN has an approximately 18 % smaller lattice constant compared to AlN [7]. Hence, strain and TDD can be reduced which normally negatively affect the efficiency of the optoelectronic device. Lattice matching also leads to reduced piezoelectric fields in the active layers which also helps increasing the efficiency of the LEDs.

However, boron exhibits a poor solubility into AlGaN [8]. To overcome this problem high temperature growth up to 1400 °C will be applied. Fortunately, only 3–4 % of boron are sufficient to compensate compressive strain in $Al_{0.75}Ga_{0.25}N$ grown on AlN.

Therefore, we recently have started a project which will clarify whether boron incorporation helps to reduce the strain in AlGaN hetero structures without deteriorating the crystal quality. Only little information about $Al_xB_yGa_{1-x-y}N$ can be found in literature [9–11]. Many fundamental properties like band gap as a function of the composition

and donor/acceptor ionization energies are still unknown or uncertain. Here, we report about our first steps to optimize the growth of AlN layers on sapphire which will be used as templates for our later AlBGaN growth studies.

2. Experimental Details

The MOVPE reactor used for this study is a low-pressure horizontal reactor (Aixtron AIX-200/4 RF-S) with a high-temperature susceptor kit enabling growth temperatures up to 1400 °C. All group III-nitrides are grown on standard (0001) sapphire substrates with an offcut of 0.3° towards the m-plane. Trimethylaluminium (TMAl), trimethylgallium (TMGa), trimethylindium (TMIn), triethylboron (TEB), and ammonia are used as precursors.

For characterization, a high resolution X-ray diffractometer (Bruker Discover D8) is used to study the crystal quality of the samples.

Photoluminescence spectroscopy is performed by using an argon fluoride excimer laser as excitation source which operates at a wavelength of 193 nm.

3. Growth of the AlN Template

Thapa and Qi [12, 13] have already investigated the growth of AlN layers and optimized the growth parameters.

Hence, we started our investigations by following their proposed recipes [14]: AlN templates are grown in a two-step process. First, an oxygen doped approximately 20 nm thick AlN nucleation layer is grown. This is followed by a faster grown undoped AlN layer with a mixture of carrier gases $N_2 : H_2 = 2 : 1$. After nucleation layer growth, a 10 min low temperature annealing step is employed at 500 °C. The TMAl and ammonia mass flows during layer growth are 10 µmol/min and 10 mmol/min, respectively which results in a V/III ratio of approximately 1000. The reactor pressure is set to 35 hPa and the surface temperature is set to 1085 °C which is measured pyrometrically from the top.

It was found that the addition of TMIn during growth helps improving the adsorption and surface mobility of the Al-precursors and hence enhances the crystal quality [12]. Due to the high growth temperature, indium is not incorporated effectively into the AlN layer [15].

XRD results for a 500 nm thick layer show very narrow reflections with a full width at half maximum (FWHM) of 92 arcsec and 1163 arcsec for the (0002) and $(10\bar{1}2)$ peaks, respectively.

Figure 1 shows the XRD measurement of the $(10\bar{1}2)$ reflection with a pseudo-Voigt regression.

The TDD can be roughly estimated by [16]

$$D = \frac{\beta}{4.35 \cdot b} \quad , \tag{1}$$

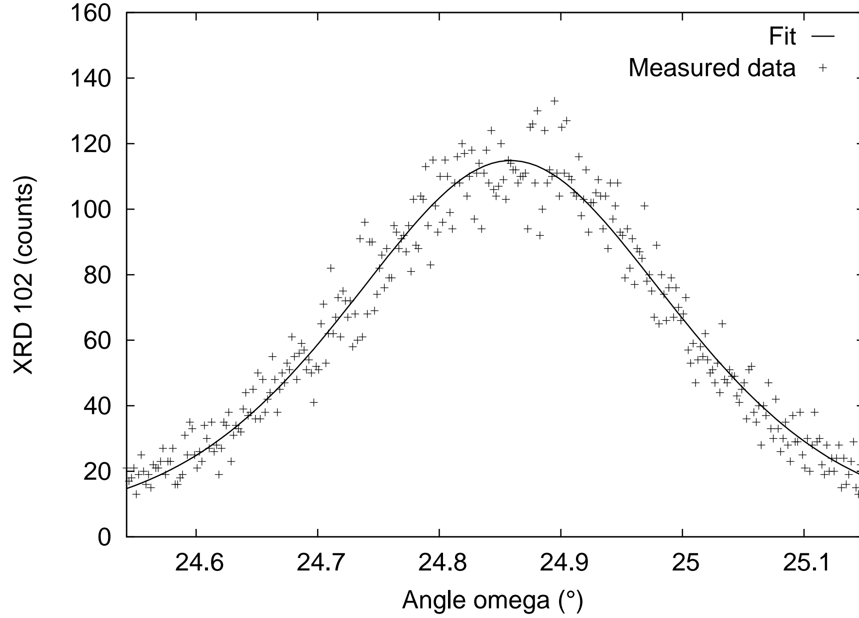

Fig. 1: XRD measurement of the $(10\bar{1}2)$ reflection of a 500 nm thick AlN template with a FWHM of 1163 arcsec.

where

$$\beta = \beta_{\text{hkl}} = \sqrt{(\beta_{\text{screw}} \cdot \cos\chi)^2 + (\beta_{\text{edge}} \cdot \sin\chi)^2} \tag{2}$$

stands for the FWHM of the respective hkl-reflection measured by XRD and b is the length of the Burgers vector of the screw- or edge-type TD, respectively. The angle χ is taken from the position of the hkl-reflection peak of the XRD's Euler cradle. From the FWHM of the (0002) reflection β_{screw} can directly be calculated since $\chi = 0$. For the calculation of the edge-type TDD the FWHM of the $(10\bar{1}2)$ reflection as well as the already obtained value for β_{screw} are used. Hence, on our recent 500 nm AlN templates we can estimate the TDDs to $D_{\text{screw}} = 2 \cdot 10^7\,\text{cm}^{-2}$ and $D_{\text{edge}} = 1 \cdot 10^{10}\,\text{cm}^{-2}$.

However, simulations show that in AlGaN MQW LEDs a TDD in the range of $10^{10}\,\text{cm}^{-2}$ is too high to achieve an internal quantum efficiency larger than a few percent [2]. Therefore, we need to reduce particularly the FWHM of the $(10\bar{1}2)$ reflection.

There are still possibilities for improvement mentioned in literature. Hirayama et al. suggest a multi-step process with faster and slower growth rates. Also AlGaN-AlN-multilayers are used to enhance the crystal properties of the template [2]. Since it is possible to install a high temperature setup in our MOVPE, higher growth temperatures will be also investigated. Pulsed growth is also successfully applied to enhance the surface mobility of the Al precursors and hence improve the crystal quality [17]. A very promising method of drastically enhancing the crystal properties is thermal annealing of the AlN template up to 1600 °C [18]. Some groups also improved the crystal quality of the AlN template by an epitaxial lateral overgrowth (ELO) process [19]. All of these approaches will be investigated to get the best possible foundation for further research.

3.1 Modification of the precursor usage

For the slow growth of the AlN:O nucleation layer, a TMAl bubbler at a temperature of 5 °C is used. This provides decently low mass flow to achieve thin and controlled nucleation layer growth. However, for the growth of the top layer, a higher growth rate is needed which requires a larger mass flow of the TMAl precursor. For that purpose, a second TMAl bubbler at a temperature of 17 °C is applied.

For the growth of boron containing materials, an additional bubbler needs to be installed into our MOVPE. Unfortunately, no spare channel is available. Therefore, one of the TMAl bubblers needs to be replaced. The idea was to find a compromise between slow and fast growth rate resulting in a bubbler temperature of 10 °C.

In order to determine which bubbler is better suited for this purpose, we have grown with different bubbler configurations, as displayed in Table 1.

Table 1: Comparison of the the crystal quality obtained with the two TMAl bubblers for a 500 nm thick AlN layer on sapphire.

	sample	T6028Aa	T6029Aa	T6033Aa
nucleation	bubbler	1	1	2
	bubbler temperature (°C)	4	4	10
	pressure (mbar)	1000	400	1400
	source flow (sccm)	25	10	25
layer	bubbler	2	1	2
	bubbler temperature (°C)	17	4	10
	pressure (mbar)	1000	400	1000
	source flow (sccm)	25	22	38
	FWHM $(10\bar{1}2)$	1500	4200	2100

The pressure as well as the temperature of the bubblers are chosen to keep the TMAl mass flow constant. Therefore, all templates are expected to have the same crystal quality. However, as can be clearly seen, the FWHM of the $(10\bar{1}2)$ reflection changes significantly depending on the bubbler used as precursor source for AlN growth. Neither bubbler one nor bubbler two can be used for both, nucleation and buffer layer growth to achieve the same FWHM as compared to the combination of both. The reason for that behaviour is not known until now. It is supposed that bubbler one might be contaminated since it is a rather old source. However, this contamination seems to help for the growth of a good nucleation layer since the crystal quality is better compared to templates with nucleation layers grown with the second bubbler. Further studies are needed to clarify this issue.

4. Summary

First studies on the growth of AlN templates were performed. Templates with a thickness of 500 nm show a FWHM of the XRD of 92 arcsec and 1162 arcsec for the (0002) and $(10\bar{1}2)$ peaks, respectively. This is equal to a TDD of $D_{\mathrm{screw}} = 2 \cdot 10^7\,\mathrm{cm}^{-2}$ and $D_{\mathrm{edge}} = 1 \cdot 10^{10}\,\mathrm{cm}^{-2}$. However, for the growth of highly efficient UV-LEDs further improvement needs to be achieved.

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