Growth and investigation of m-plane (In)GaN buffer layers on LiAlO₂ substrates

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We deposited pure m-plane GaN (1-100) layers on LiAlO₂ (100) substrates by MOVPE using Mg-doped InGa_N buffer layers of various thickness. These sealing layers are grown under nitrogen ambient and help to improve the film coalescence while reducing the oxygen background doping of the GaN films. We show that for an increasing thickness of the InGa_N:Mg buffer layer, the highly strained GaN layer slightly relaxes. An improved surface morphology can be achieved for thicker InGa_N:Mg buffers while the defect density clearly increases above a certain thickness as it can be seen in X-ray rocking curve measurements. In temperature-dependent photoluminescence spectroscopy, we observe not only peaks due to DAP transitions at an energy of around 3.3 eV, but also a defect related peak at 3.43 eV as well as a peak at 3.49 eV connected to free and bound excitonic transitions. It is interesting to note that for an increasing thickness of the InGa_N:Mg buffer, we detect a higher intensity of the excitonic transition while the defect-related signal seen at 3.43 eV is getting more pronounced. It might be possible that during the film relaxation, basal plane stacking faults are introduced into the layer.

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1 Introduction

The increasing demand for highly efficient nitride blue and green light emitting diodes (LEDs) has attracted great interest for the growth of nonpolar GaN. In contrast to the common growth of nitride material in the polar c-axis, the absence of polarization-induced electric fields in the growth direction for nonpolar structures leads to a better overlap of electron and hole wave functions in quantum wells and therefore to a more efficient carrier recombination [1]. As a second advantage, nonpolar light emitters exhibit a much lower wavelength shift with higher driving currents due to the absence of polarization field screening normally occurring in c-plane GaN [2]. One interesting approach to achieve this is to grow on (100) LiAlO₂ substrates, which enables a deposition of GaN in m-plane orientation with a rather small substrate-lattice mismatch of only 1.7 % and 0.3 % in [11-20] and [0001] direction of GaN, respectively [3]. The substrates can be manufactured by conventional Czochralski pulling method which allows potentially cheap large-size wafer production. One of the challenges with this material is the sensitivity to hydrogen and water. On the other hand, this offers a possibility of an easy substrate removal after device processing for an improved light extraction and heat management.

In first literature reports, mainly molecular beam epitaxy (MBE) was used to deposit pure m-plane GaN on LiAlO₂ [1,3,4] but also successful growth by hydride vapor phase epitaxy (HVPE) has been reported [5]. The challenges with the growth by metal organic vapor phase epitaxy (MOVPE) on this substrate are related to the demand for high growth temperatures and hydrogen ambient to achieve high-quality material, both of which can cause serious substrate damage. Nevertheless, several groups succeeded in growing GaN on LiAlO₂ by MOVPE [6,7]. But the layers still exhibit either a low phase purity, rough surfaces or a high background carrier concentration, the latter being caused by an outdiffusion of oxygen from the substrate at elevated temperatures. As we have shown in an earlier report, the introduction of a thin layer of Mg-doped InGa_N helps to effectively seal the substrate surface and to block the outdiffusion of oxygen into the layer [8]. By using this approach of growing m-plane GaN, nonpolar MQW structures with high wavelength stability could be produced [9].

To study and improve this method, we investigated m-plane GaN buffers using InGa_N:Mg sealing layers of different thicknesses and their influence on the defect characteristics of the GaN films.

Experimental

The growth experiments were performed in single-wafer, horizontal-flow AIXTRON MOVPE reactors equipped with TMGa, TMIn and Cp_2Mg as precursors. Polished (100) LiAlO_2 substrates with an average substrate root mean square (RMS) roughness of 0.4 - 0.5 nm were used without any cleaning procedure. More details about their fabrication can be found elsewhere [10]. The process was initiated with an in-situ nitridation by exposing the wafer to NH_3 at 1050°C . The Mg-doped InGaN sealing layer with a thickness between 25 and 150 nm was deposited at 900°C . This was followed by a 200 nm thick Mg-doped GaN film at 1000°C and a 500 nm thick undoped GaN layer grown at 1050°C . Phase purity and crystal quality of the m-plane GaN were attested by high resolution X-ray diffraction (HRXRD) using a PANalytical Material Research Diffractometer and software analysis tool. The evaluation of strain and indium fraction was performed using methods and elastic constants given in [11]. Atomic force microscopy (AFM) scans were performed to measure the surface roughness. To determine optical properties, we carried out temperature-dependent photoluminescence spectroscopy (PL) using a He closed-cycle cryostat and a 325 nm HeCd Laser with an excitation power density of $\sim 1 \text{ W/cm}^2$. Hall measurements were conducted in van der Pauw geometry.

Results and discussion

To prevent damaging of the sensitive LiAlO_2 surface, we used a relatively low reactor temperature and nitrogen ambient in the first growth step. As a starting layer, we chose Mg-doped InGaN since this material can be grown under nitrogen ambient at reasonable quality. The Mg doping serves the purpose to block oxygen from diffusing out of the substrate which will otherwise lead to a very high n-type background carrier concentration in the GaN layer [8,12]. In Fig. 1, HRXRD $2\theta/\omega$ scans in triple axis geometry of four InGaN:Mg/GaN structures having various InGaN:Mg buffer thicknesses are displayed. Beside of the substrate peak at 34.69° , we can observe peaks for m-plane GaN and m-plane InGaN while no c-plane material is detected. With rising InGaN:Mg thickness, we see an increasing intensity of the correlated peak. The peaks for GaN are located around 32.27° , which is similar to what other groups report [1,3]. But for the thickest InGaN:Mg buffer, this peak shifts to 32.32° which is closer to its ideal position at 32.39° . This can be explained by a relaxation of the strained GaN film, possibly supported by the underlying InGaN:Mg layer. Reciprocal space maps (not shown here) around (2-200), (2-310) and (2-20-1) reflections on the sample with 25 nm InGaN:Mg revealed an In molar fraction of 3.4% and an in-plane compressive strain for the GaN layer of -1.1% and -0.1% in [11-20] and [0001] direction, respectively. These values are consistent with the lattice mismatch and other reports about MBE-grown m-plane GaN layers on LiAlO_2 [4,13]. In contrast to that, the sample with the thickest InGaN:Mg layer features an In fraction of 7.6% and strain values of -0.5% and 0.1% for the [11-20] and [0001] direction, respectively, indicating a relaxation of the GaN film. The InGaN:Mg film is also strained and exhibits a slight shift to lower angles with rising In content in (2-200) $2\theta/\omega$ scans (not shown here). Because of the superposition of the InGaN and GaN (1-100) peaks, the shift cannot be observed in Fig. 1. We ascribe this increase of the In concentration to the well-known indium pulling effect [14].

In order to investigate the influence of the InGaN:Mg layer and its thickness on the GaN material quality, X-ray rocking curve (XRC) measurements with the in-plane component of the incident beam along GaN [0001] and AFM scans for a $5 \times 5 \mu\text{m}^2$ area were performed on all samples. The direction of the incident beam was chosen parallel and perpendicular to the c-axis to uncover anisotropic defect influences. In Fig. 2, both the XRC full width at half maximum (FWHM) values and the AFM root mean square (RMS) roughness values are shown. The surface morphology shown in the inset of Fig. 2 for a 25 nm (a) and 150 nm (b) thick InGaN:Mg buffer exhibits trenches indicating a not fully coalesced surface. The average surface roughness clearly improves with the thickness of the underlying InGaN:Mg coating which is possibly caused by a better layer coalescence in the first growth steps. Apart from that, we see an increase of the FWHM values parallel to the c-axis above a thickness of 50 nm, while the values measured in perpendicular direction show a slightly oppositional trend. As in the former geometry the measurement should become more sensitive to basal plane stacking faults (BSF) [15], this indicates a possible increase of such defects connected to the strain relaxation for thicker InGaN:Mg films.

To obtain more information on the defect characteristics in our layers, we employed temperature-dependent PL spectroscopy. Fig. 3 shows the PL spectra of a GaN layer with 50 nm InGaN:Mg taken in the temperature range from 10 to 300 K. For low temperatures, at least two strong peaks can be seen for energies between 3.1 and 3.3 eV, which we think might origin from DAP transitions due to the oxygen background concentration and residual Mg doping [16,17]. Furthermore, we observe a high signal at 3.43 eV, which has been reported to be the energetic position for stacking faults in m- and a-plane GaN [18,19]. While this signal is very high for low temperature, it is quenched quickly with temperature increase and disappears from the spectrum at 120 K. Another third peak is visible in Fig. 3 around 3.49 eV. We observe a complicated temperature dependence of the position of this peak, namely a high-energy shift for 10 - 60 K and a low-energy shift at temperatures higher than 60 K. Such complex behaviour is caused by the superposition of the bound and free excitonic (BE+FE) signal in one broad peak. We can see that the signal from bound excitons determines the energetic peak position for very low temperatures. At temperature enhancement, the peak shifts smoothly from bound to free exciton position which dominates the peak for temperatures higher than 70 K. It is also interesting to note that with increasing thickness of the InGaN:Mg layer, we observe a clear tendency for the intensity ratio of the excitonic and defect-

related transitions. This trend is illustrated in Fig. 4. While the BSF-related peak at 3.43 eV shows an increasing intensity for thicker InGa_N:Mg layers, the signal at 3.49 eV is getting weaker. We think this may be caused by an increase of the stacking fault density in the m-plane GaN layers.

Conclusion

We have deposited and characterised m-plane GaN films grown by MOVPE using Mg-doped InGa_N buffer layers of various thicknesses. It was shown that the thickness of this starting layer affects the properties of the overlying GaN film. Thicker InGa_N:Mg layers lead to a distinct relaxation of the highly strained GaN. Furthermore, thick buffer layers produce an improved surface morphology. However, we also observe a decrease in crystal quality, possibly caused by an increased BSF density. Optical investigations yield also a peak at 3.43 eV which is present in all of our samples and can be ascribed to BSF-related transitions. The higher intensity of this peak for thicker InGa_N:Mg layers confirms our statement about the rising defect density. We also find a signal from DAP transitions in our layer, which is due to the still high concentration of oxygen impurities and some residual Mg doping. Beside of that, another peak at 3.49 eV which is connected to both free and bound excitons is visible already indicating quite sufficient quality of our layers.

Acknowledgments

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Figures

Figure 1 $2\theta/\omega$ scans of InGaN:Mg/GaN buffers on LiAlO₂ with varying InGaN:Mg thickness. The dotted line on the left side is a guide to the eye. The dashed line shows the unstrained position of m-plane GaN.

Figure 2 Development of the (1-100) XRC FWHM with the in-plane component of the incident beam parallel ([0001], closed triangles) and perpendicular ([1-120], open triangles) to the c-axis and AFM roughness for various InGaN:Mg buffer layer thicknesses. The inset shows AFM surface scans for the samples with 25 nm (a) and 150 nm (b) thick InGaN:Mg buffer.

Figure 3 Temperature-dependent PL spectra for an m-plane GaN structure with 50 nm InGaN:Mg buffer.

Figure 4 Low-temperature PL signal intensity ratios of both excitonic and defect-related peaks versus InGaN:Mg thickness.