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PEMBE-Growth of Gallium Nitride on (0001) Sapphire: A comparison to MOCVD grown GaN

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Abstract

Thin films of GaN on c-plane sapphire were grown by plasma-enhanced molecular beam epitaxy (PEMBE). The influence of different growth conditions on the quality of the epitaxial layers was studied by x-ray diffraction (XRD), atomic force microscopy (AFM) and Hall measurements. For low deposition temperatures, the growth of a thin buffer layer of AlN results in a decrease of the XRD rocking curve full width at half maximum (FWHM) but also in poorer quality in electronic and optical properties. Samples of 3µm thickness with 570 arcsec FWHM in the XRD rocking curve, a near band gap PL-emission FWHM at 5 K of 7 meV, charge carrier densities of $n_e = 2 \times 10^{17}$ cm^{-3}, and Hall mobilities of 270 cm^2/Vs at 300 K were grown without a buffer layer. A comparison of the morphology and XRD rocking curves with those of GaN films deposited by metalorganic chemical vapour deposition (MOCVD) shows that the two methods have different growth mechanisms.

1. Introduction

Metalorganic chemical vapour deposition (MOCVD) and molecular beam epitaxy (MBE) are the most common methods for epitaxial growth of GaN films. Both techniques have their advantages. The MOCVD is a comparatively inexpensive method, because ultrahigh vacuum is not necessary. The MBE benefits from the clean environment and therefore low concentration of unwanted extrinsic defects, excellent control of the deposition parameters, and lower deposition temperatures which should lead to fewer thermally induced intrinsic defects. The plasma-enhanced molecular beam epitaxy (PEMBE) uses nitrogen radicals, produced in a nitrogen rf-plasma. Hydrogen, probably responsible for the passivation of dopants such as magnesium [1], is thus avoidable during PEMBE growth in contrast to MBE using other nitrogen sources [2]. Despite these advantages, the best film quality has so far been achieved by the MOCVD method [3]. In this paper we discuss the PEMBE growth of GaN and emphasize some differences in MOCVD and PEMBE-grown GaN.

2. Experimental

The PEMBE GaN films are grown in a Tectra Mini MBE® chamber with a background pressure of 1×10^{-10} mbar . The gallium and aluminum sources are conventional hot lip and cold lip effusion cells, respectively. As nitrogen source, an Oxford Applied Research CARS25 radical beam source is used. In this source the molecular nitrogen is decomposed in a radio frequency (rf) plasma at 13.56 MHz. The activated nitogen is predominantly atomic nitrogen and has only a negligible fraction of ions [4]. This is the main difference to the electron cyclotron resonance (ECR) plasma source, [4] the other commonly used source for PEMBE growth of III-V nitrides. During the growth, the samples were rotated with 10 rpm to obtain uniform films over 2 inch (0001) sapphire wafers. Before deposition the substrates were pretreated by heating for 1 hour at 950 °C under N-radical flux. The deposition temperatures were varied from 650°C up to 825°C. The Ga flux onto the (0001) sapphire surface was 8×10^{14} cm^{-2}s^{-1} as measured with a quartz microbalance. The pressure in the MBE chamber during deposition and pretreatment was 4×10^{-5} mbar. The inductively coupled rf power was 400W. Before the actual GaN growth, on several samples, AlN buffer layers of approximately 50 nm thickness were grown at 850 °C with an Al flux of 6×10^{13} cm^{-2}s^{-1}. To ensure that all of the Al metal on the wafer surface reacts to AlN, the N-radical flux was further maintained for 30 min.

The MOCVD samples were grown by low pressure MOCVD [5], using ammonia and triethylgallium (TEG) as precursors for nitrogen and gallium at a temperature of 1020 °C on a 50 nm thick GaN buffer layer produced at 520 °C. The ammonia flux was 1.8 slm and hydrogen and nitrogen fluxes were 0.7 slm each. The flow rate of bubbled hydrogen was 10 sccm, yielding a flow rate of 1.8 μmol/min TEG in the reaction chamber. The resulting deposition rate was 1 μm/h. Typical concentrations of free carriers in 1 μm thick GaN films were 1×10^{17} cm^{-3} with a mobility of 100 cm^2/Vs.

The XRD rocking curve measurements were performed with a single monochromatized two-circle diffractometer. The carrier concentration and mobility were characterized by Hall measurements, using the van der Pauw technique.

3. Results and discussion

For optimizing the film quality of MBE-GaN, several deposition parameters, such as Ga flux, buffer layer growth, and deposition temperature were varied. To prevent the formation of Ga droplets at low deposition temperatures, the Ga flux was set at 3×10^{14} $cm^{-2}s^{-1}$ (Figure 1) corresponding to a Ga effusion cell temperature of 950 °C.

At a substrate temperature of 775°C, an increase in Ga flux to 8×10^{14} $cm^{-2}s^{-1}$ leads to a proportional increase in deposition rate. This fact, and a significant decrease of the growth rate by elevating the deposition temperature, are indicative of Ga-limited growth. An increase of the Ga flux for a given temperature also affects the electronic properties of the material. The Hall mobility remains at 35 cm^2/Vs, but the carrier concentration decreased from 4×10^{18} for low flux to $n_e = 1 \times 10^{17}$ cm^{-3} for elevated flux. This is evidence for reduced intrinsic defect density and therefore for improved stoichiometry. The structural quality does not change significantly, as shown by the XRD rocking curves with FWHM of 0.49° for both samples. The dependence of film quality on substrate temperature for GaN without an AlN buffer layer is shown in Figure 2. The increasing Hall mobility for constant charge carrier density, $n_e = 2 \times 10^{17}$ cm^{-3}, and the decrease in the XRD rocking curve FWHM is evidence for improvement, particularly in the structural properties of the epitaxial GaN.

The improvement of film quality by the deposition of a thin GaN or AlN buffer layer is well documented for MOCVD [6] [7]. The growth of approximately 50 nm thick AlN buffer layers in MBE leads, especially at low deposition temperatures, to a reduced mosaicity of the GaN film, which is mainly responsible for the FWHM in the XRD rocking curves for symmetric reflections. Though no significant change in morphology occurred, nevertheless a drastic change in optical and electronic properties of the films was observed. The resistivity increased from about 10^3 Ωcm for GaN films without a buffer layer to the order of 10^9 Ωcm for samples with an AlN buffer layer, thus preventing Hall measurements. Though the FWHM of the XRD rocking curve is not strongly affected by the buffer layer, a strikingly increased background due to diffuse scattering was measured for samples prepared at elevated deposition temperatures (Figure 3). This indicates that an AlN buffer layer has negligible influence in the growth direction [00.1], but strongly affects the formation of defects in other directions. PL measurements showed that the near-bandgap luminescence disappears for samples grown with an AlN buffer layer. This also points to the formation of deep defects that provide nonradiative recombination paths.

In order to investigate the role of thickness on the film properties, a 3 μm thick sample was grown at 810°C with Ga flux of 8×10^{14} $cm^{-2}s^{-1}$. Compared with a 1 μm thick film, the Hall mobility increased from 135 to 270 cm^2/Vs for the same charge carrier density of 2×10^{17} cm^{-3}, and the FWHM of the XRD rocking curve decreased from 840 to 570 arcsec. This large influence is assumed to be due to decreasing defect density, particulary the reduction of grain boundaries, with increasing film thickness.

MOCVD samples with a similar rocking curve FWHM as the PEMBE films show less diffuse scattering, for the same sample thickness. This fact, and more directly a comparison of the morphology of PEMBE and MOCVD films (Figure 4), show that different growth mechanisms dominate. A three-dimensional growth for PEMBE films leads to epitaxial islands, responsible for a high surface roughness of about 30 nm and probably to abrupt interfaces between the crystallites. For the MOCVD samples, two-dimensional growth is achieved as indicated by smooth surfaces with a roughness of 5 nm, corresponding to 20 monolayers over 10 μm.

4. Conclusions

Several PEMBE deposition parameters were investigated for their influence on sample quality. An increase in Ga flux from 3 to 8×10^{14} $cm^{-2}s^{-1}$ leads to a lower charge carrier density from $n_e = 4 \times 10^{18}$ to 1×10^{17} cm^{-3}

respectively. In contrast to the growth of GaN by MOCVD, the use of AlN buffer layers did not improve the sample quality. The Hall mobility and the structural quality can be improved by the growth of thick samples. Comparing the morphology of PEMBE and MOCVD samples indicates that the mechanism is two-dimensional growth for MOCVD and three-dimensional growth for PEMBE, leading to a high concentration of inner interfaces. Nevertheless, GaN films with a Hall mobility μ_H = 270 cm^2/Vs , charge carrier density n_e = 2 x 10^{17} cm^{-3} and XRD rocking curve FWHM of 570 arcsec are well suited for device fabrication by PEMBE.

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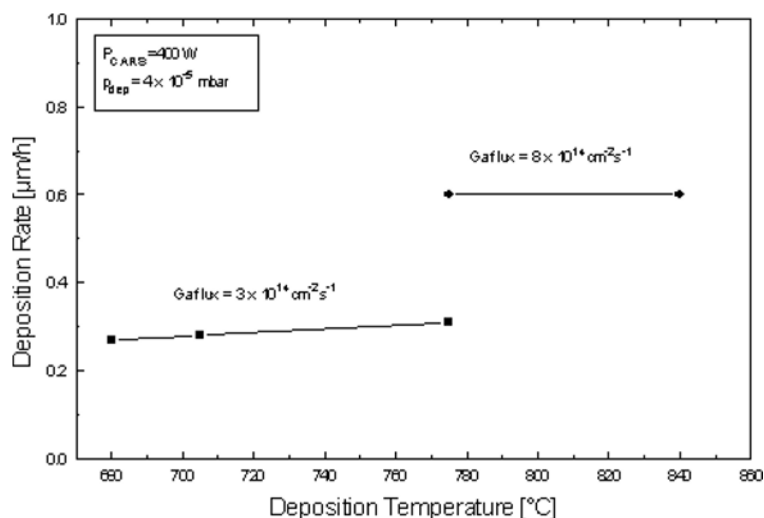

Figure 1. Dependence of the deposition rate on substrate temperature and Ga flux.

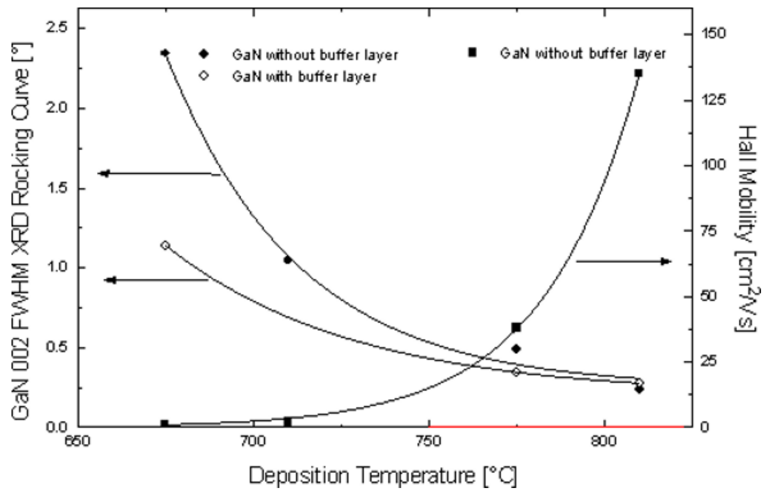

Figure 2. The FWHM of the XRD rocking curve and the Hall mobility at 300K versus the substrate temperature for MBE grown samples. All GaN films have similar thickness (0.7 - 1.1 µm) and, for samples without a buffer layer, similar charge carrier densities (1 - 3 x 10^{17} cm^{-3}).

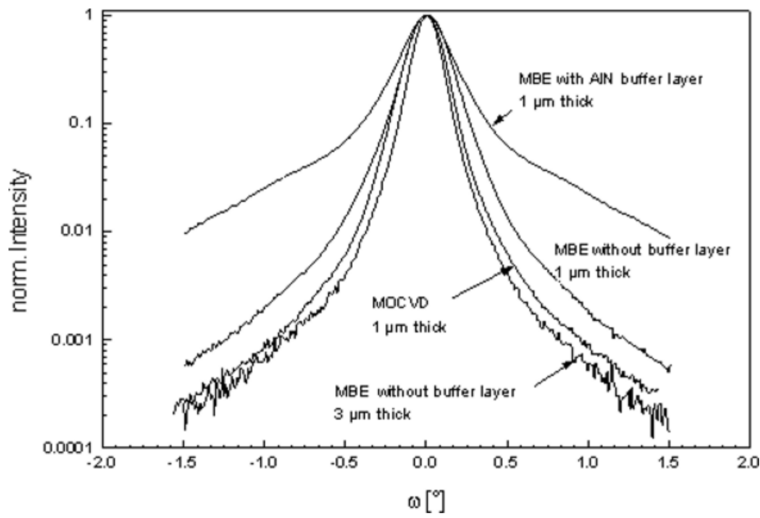

Figure 3. XRD rocking curves for MBE samples produced with the same parameters (T_{dep}= 810 °C, Ga flux = 8 x 10^{14} $cm^{-2}s^{-1}$), on a logarithmic scale. For comparison, a MOCVD sample with similar FWHM is shown.

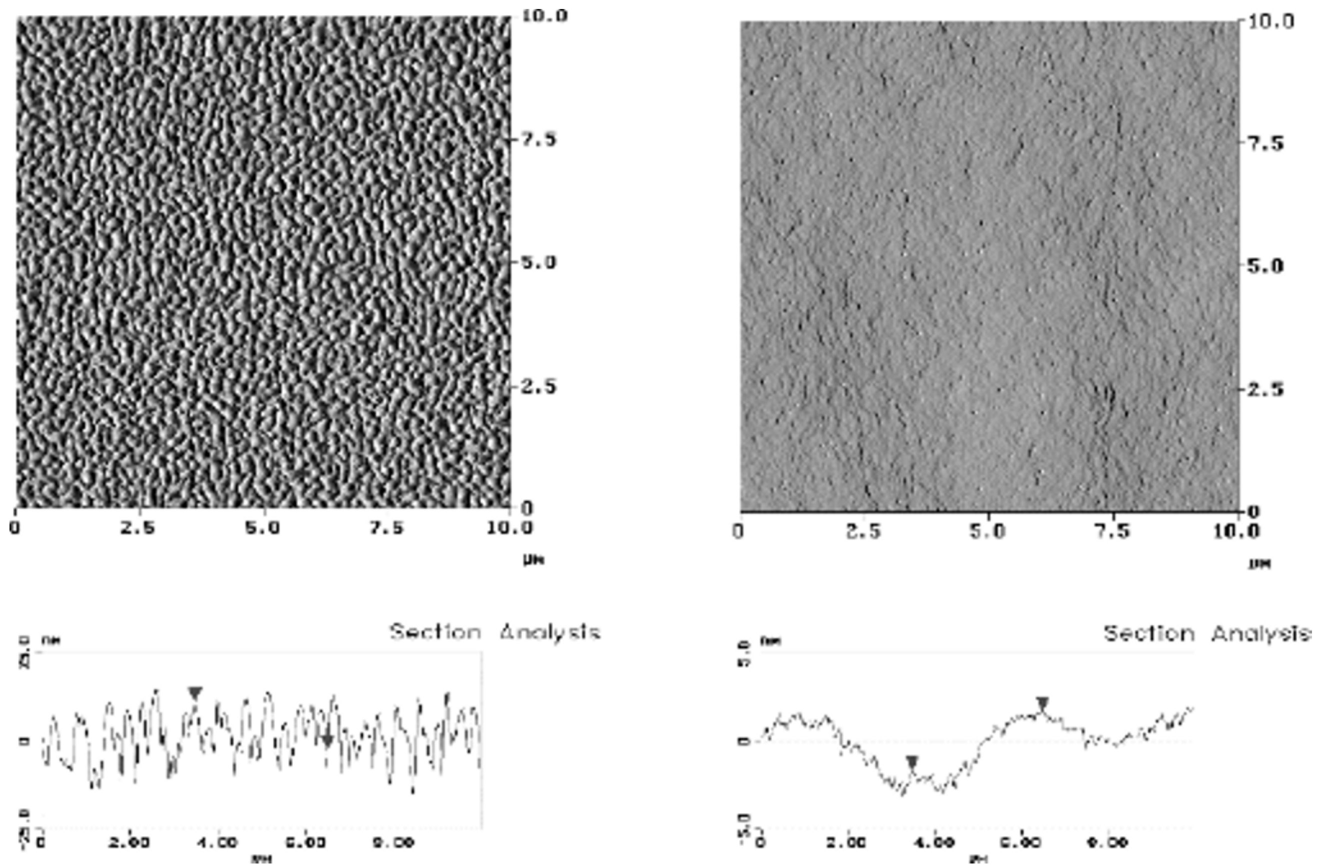

Figure 4. AFM surface image and depth profiles of PEMBE (left) and MOCVD (right) grown GaN with similar thickness and XRD rocking curve FWHM.

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