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Growth Of High Quality GaN Thin Films By MBE On Intermediate-temperature Buffer Layers

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We report the growth of high quality GaN epitaxial layers by rf-plasma MBE. The unique feature of our growth process is that the GaN epitaxial layers are grown on top of a double layer that consists of an intermediate-temperature buffer layer (ITBL), which is grown at 690°C and a conventional low-temperature buffer layer deposited at 500°C. It is observed that the electron mobility increases steadily with the thickness of the ITBL, which peaks at 377 $cm^2V^{-1}s^{-1}$ for an ITBL thickness of 800 nm. The PL also demonstrated systematic improvements with the thickness of the ITBL. Our analyses of the mobility and the photoluminescence characteristics demonstrate that the utilization of an ITBL in addition to the conventional low-temperature buffer layer leads to the relaxation of residual strain within the material resulting in improvement in the optoelectronic properties of the films. A maximum electron mobility of 430 $cm^2V^{-1}s^{-1}$ can be obtained using this technique and further optimizing the growth conditions for the low-temperature buffer layer.

1 Introduction

Various groups have attempted different techniques for improving the electrical and optical properties of MBE grown GaN thin films. The nitrogen source is generally provided by an ECR source, an rf-plasma source or by gaseous NH_3. It was reported by Tang *et al*. [1] that electron mobility, up to 560 $cm^2V^{-1}s^{-1}$, can be obtained using a UHV magnetron sputtered AlN buffer layer and a epitaxial GaN layer grown using NH_3 as the nitrogen source. High quality GaN was also grown using a GaN template deposited by migration enhanced epitaxy in conjunction with an AlN/GaN superlattice layer. A mobility of 668 $cm^2V^{-1}s^{-1}$ was reported using this growth technique [2]. Recently, Heying *et al.* [3] reported the highest mobility to date of 1191 $cm^2V^{-1}s^{-1}$ grown by rf-plasma assisted MBE on MOCVD-GaN/sapphire composite substrates.

2 Experiment And Results

In this paper we report the growth of high quality GaN epilayers by MBE on (0001) oriented sapphire wafers. The activated nitrogen is provided by an EPI UNI-Bulb rf-plasma source. The substrate was first cleaned using a standard cleaning procedure. It was then outgassed at 800°C, followed by a nitridation process within the growth chamber at 500°C. A conventional low-temperature buffer layer of thickness about 20 nm was grown at 500°C. An intermediate-temperature buffer layer (ITBL) was then grown on top of the conventional buffer layer at 690°C. The thickness of the ITBL was varied up to 1.25 µm. Finally, a 1.8 µm layer of high temperature, slightly n-doped GaN epitaxial layer was grown at 750°C. The surface morphology and optimal III/V ratio were monitored *in-situ* by the reflection high-energy electron diffraction (RHEED) pattern. The typical electron beam voltage used was 9 kV. All samples exhibited (1×1) RHEED patterns during growth. It was found that upon cooling down below 300°C, (3×3) reconstructed RHEED patterns were seen for all samples and a typical RHEED image for the sample grown on a 800-nm-thick ITBL is shown in Figure 1.

The optoelectronic properties of the GaN epitaxial layers were characterized by Hall and photoluminescence (PL) measurements. The room temperature Hall coefficient was measured by the Biorad HL5500 system. From the Hall coefficients the carrier concentrations for the various films were found to be around 3×10^{17} cm^{-3}. The electron mobilities for the films were also evaluated. The experimental results are shown in Figure 2. Most interestingly, the electron mobility is found to increase steadily with the thickness of the ITBL. Typical mobility for films grown without an ITBL is 87 $cm^2V^{-1}s^{-1}$ and the maximum mobility recorded for this series of samples is 377 $cm^2V^{-1}s^{-1}$ for films grown with an ITBL thickness of 800 nm. Further increase in the ITBL thickness beyond 800 nm results in the gradual degradation in the mobility. For an ITBL thickness of 1.25 μm, the electron mobility is found to be 355 $cm^2V^{-1}s^{-1}$.

The room temperature PL of the films were characterized systematically. In Figure 3 we show typical PL spectra for a film grown without an ITBL and one grown with an ITBL thickness of 800 nm. From the data we observe that the spectra exhibit very low yellow emission, indicative of high structural quality for both films. Detailed analyses of the PL spectra show that the magnitudes of the PL spectra also increase systematically, following the same trend as the mobility, as a function of the ITBL thickness. From Table 1 we observe that the magnitude of the PL spectrum for the film grown with an ITBL thickness of 800 nm is found to be increased by a factor of 2.3 compared to the films grown without an ITBL. In addition, it is noteworthy that the near band edge peak positions, λ_p, of the PL spectra are observed to vary systematically as well. The typical results are summarized in Table 1. From the data we observe that λ_p is 367.2 nm for the film grown without an ITBL. As the ITBL thickness increases, λ_p is found to decrease systematically. For an ITBL thickness of 800 nm, λ_p = 366.5 nm. Further increase in the ITBL thickness leads to a rebound phenomenon in λ_p, and for an ITBL thickness of 1.25μm λ_p = 367.0 nm.

3 Discussions

The experimental data clearly demonstrate that the quality of the GaN thin films improves steadily with the thickness of the ITBL. Such improvement cannot be explained simply by the increase in the total thickness of the GaN films. As a control experiment, a GaN epilayer of thickness equal to 2.6 μm was grown without an ITBL; all other experimental conditions were identical to the films grown with ITBL. The mobility of the film is found to be about 170 $cm^2V^{-1}s^{-1}$. This clearly shows that ITBL plays a significant role in the improvement of the film quality.

The pronounced (1×1) streaks seen at the growth temperature, 750°C, suggested that the GaN surface is unreconstructed during growth. It is believed that the (1×1) unreconstructed surface is due to a monolayer of Ga which is tightly bound to the GaN [4]. All samples in this study are N-face as suggested by the (3×3) reconstructed RHEED patterns upon cooling down below 300°C. In addition, the N-face characteristic is further confirmed by etching in molten KOH solution.

The significant improvement in the carrier mobility is attributed to the reduction in threading dislocations. It has been shown that edge dislocations introduce acceptor centers along the dislocation lines, which capture electrons from the conduction band in an *n*-type semiconductor [5]. The dislocation lines become negatively charged and a space charge is formed around it, which scatters electrons traveling across the dislocations and as a consequence, the electron mobility is reduced. Heying *et al.* [6] demonstrated that x-ray rocking curves for off-axis reflections such as (102) plane is a reliable indicator of the threading dislocations in GaN thin films. However, x-ray analysis is not suitable in our case since the x-ray diffraction peak from the ITBL will overwhelm the full width half maximum of the measured rocking curve. Therefore, transmission electron microscopy (TEM) is one of the best ways to reveal the structural improvement by means of ITBL and a TEM experiment is underway which will be subject of a separate publication.

The correlated variations in the mobility and the PL spectra are indicative of a common mechanism behind both phenomena. The systematic shift in the peak position of the PL is attributed to the change in excitonic transition energies for the different GaN films. This results from the relaxation of residual strain in the epilayers due to the mismatches of lattice constants and coefficients of thermal expansion between GaN and the sapphire substrate [7], [8], [9]. The strain-related phenomena in GaN epitaxial films have been well investigated both experimentally and theoretically [7], [10]. A number of authors have shown that the relaxation of the residual strain is associated with the shift in the PL and photoreflectance peak positions [9], [10], [11]. Shikanai *et al*. reported that the energy of the free excitons associated with the top valence band varies linearly with the in-plane and the axial components of the strain tensor [10]. These results indicate that the band structure of GaN is strongly influenced by the residual strain. The excitonic transition energy increases under compressive biaxial strain, and decreases under tensile biaxial strain. The small excitonic transition energy for the sample grown without ITBL indicates a large tensile stress existing in the film. Our PL results show that the tensile stress relaxes with the use of ITBL. The results agree well with previous report by Kisielowski *et al*. [8], which is in contradiction to the results observed in MOCVD and HVPE grown GaN films, where the overall effects of strain generated in GaN is found to be compressive [9], [11].

The experimental results on electron mobility and PL exhibit the same dependencies on the thickness of the ITBL. From Table 1 we observe that the peak energy of the PL increases steadily with the thickness of the IBTL, indicative of the relaxation of the residual tensile strain as ITBL thickness increases. However, for the ITBL thickness beyond 0.8 µm, both the electron mobility and the PL are seen to degrade slightly. This is associated with the rebound in the peak position of the PL, indicative of the increase in the residual strain for ITBL thickness larger than 0.8 µm. The physical process that underlies this phenomenon is not known for certain at this time. It is possible that a certain critical thickness may exist for the ITBL. More systematic characterizations of the films are needed to pinpoint the mechanisms responsible for the observed degradation of the films.

It is possible that the utilization of the ITBL may affect the optimal conditions for the growth of the low-temperature buffer layer. Initial results in our laboratory show that a maximum mobility of 430 $cm^2V^{-1}s^{-1}$ can be obtained by varying the thickness of the low-temperature buffer layer while keeping the thickness of the ITBL at 800 nm. A systematic study is underway to confirm this point and to investigate the optimal conditions for the growth of the double buffer layer system. This will be subject of a separate publication.

4 Conclusion

In conclusion, we have conducted systematic investigation of the electronic and optical properties of GaN thin films grown by rf-plasma MBE. An intermediate-temperature buffer layer was first grown on top of the conventional low-temperature buffer layer before the deposition of the GaN epilayers. The thickness of the intermediate-temperature buffer layers were systematically varied up to 1.25 µm. The electron mobility is found to improve with the thickness of the intermediate-temperature buffer layer, which peaks at at 377 $cm^2V^{-1}s^{-1}$ for a thickness of 800 nm for the intermediate-temperature buffer layer. Further increase in the thickness of the intermediate-temperature buffer layer results in the gradual degradation in the electron mobility. The PL spectra are found to follow the same trend as the electron mobility. In addition, we observe a systematic shift in the peak position of the PL as a function of the intermediate-temperature buffer layer thickness with a trend that corroborates the variation of the electron mobility. Our studies show that the changes in the optical and electronic properties of the GaN films with the thickness of the buffer layer result from the corresponding variations in the residual strain in the films. Our work also shows that more work is needed to optimize the growth conditions of the double buffer layer system. Initial work in our laboratory has shown that electron mobility as high as 430 $cm^2V^{-1}s^{-1}$ can be obtained by varying the thickness of the low-temperature buffer layer.

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FIGURES

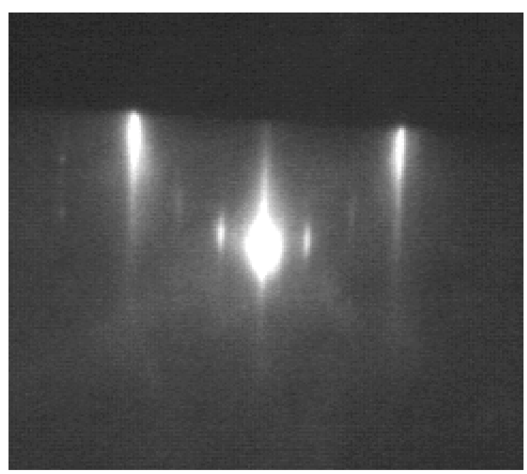

Figure 1. GaN (3 × 3) RHEED pattern observed with the electron beam along the $[2\,\bar{1}\,\bar{1}\,0]$ direction.

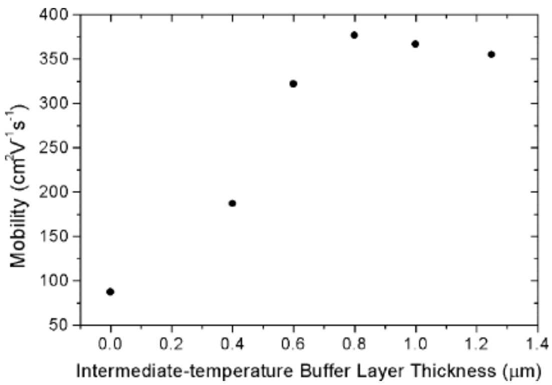

Figure 2. Room temperature mobility for different thickness of intermediate-temperature buffer layer.

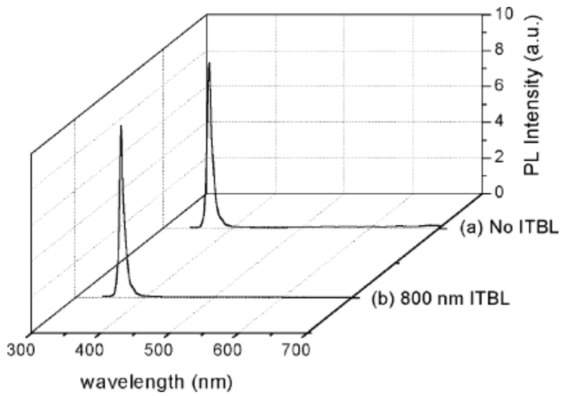

Figure 3. Typical room temperature photoluminescence spectra of GaN (a) grown without an intermediate-temperature buffer layer (ITBL) and (b) grown in presence of a 800-nm-thick ITBL.

TABLES

Table 1. Normalized intensity and peak position of room temperature photoluminescence near band edge emission for various thickness of intermediate-temperature buffer layer.

Thickness of ITBL (nm)	Normalized near band edge intensity	Near band edge peak position λ_p (nm)
0	0.44	367.2
400	0.62	367.0
600	0.79	366.8
800	1	366.5
1000	0.91	366.8
1250	0.77	367.0