GRADED InGaN BUFFERS FOR STRAIN RELAXATION IN GaN/InGaN EPILAYERS GROWN ON SAPPHIRE

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ABSTRACT

Graded InGaN buffers were employed to relax the strain arising from the lattice and thermal mismatch in GaN/InGaN epilayers grown on sapphire. An enhanced strain relaxation was observed in GaN grown on a stack of five InGaN layers, each 200 nm thick with the In content increased in each layer, and with an intermediate thin GaN layer, 10 nm thick inserted between the InGaN layers, as compared to the conventional two-step growth of GaN epilayer on sapphire. The function of the intermediate layer is to progressively relax the strain and to annihilate the dislocations that build up in the InGaN layer. If the InGaN layers were graded too rapidly, more dislocations will be generated. This increases the probability of the dislocations getting entangled and thereby impeding the motion of the dislocations to relax the strain in the InGaN layer. The optimum growth conditions of the intermediate layer play a major role in promoting the suppression and filling of the V-pits in the GaN cap layer, and were empirically found to be a thin 10 nm GaN grown at 750 ^{0}C and annealed at 1000 ^{0}C.

1. INTRODUCTION

Nitride semiconductors are currently one of the most encouraging materials for the fabrication of visible-to-ultraviolet optoelectronic devices. These semiconductors crystallize preferentially in the hexagonal (wurtzite) structure. One of the obstacles that have hindered the development of GaN is the lack of a suitable substrate material that is lattice-matched to and thermally compatible with GaN. Sapphire is the most widely used substrate, despite the very large lattice mismatch (~14%) between GaN and Al_2O_3. An intermediate low temperature nucleation AlN or GaN buffer layer is usually grown to improve the quality of the GaN layer but major structural imperfections are still found in the high temperature layer. Threading dislocation (TD) densities of about 10^8 – 10^{10} cm^{-2} are typical[1].

InN and the ternary $In_xGa_{1-x}N$ remain the most mysterious compound due to the difficulty in growing high-quality crystals due to the extremely high equilibrium vapour pressure of nitrogen. The most frequently observed defects in InN and the ternary $In_xGa_{1-x}N$ are V-pits. A V-pit has a shape of an open hexagonal, inverted pyramid that is defined by the six $\{10\bar{1}1\}$ planes[3]. Thus, in cross section this defect appears as an open ''V''. The V-pit should obey the relationship of $h = 1.63a$, where h is the depth of a V-pit and a is length of the side forming a symmetrical hexagon[3]. a shall be referred to as the radius of the hexagon hereafter. V-pits are believed to have originated from either TD[4] or generated from the stacking mismatch boundaries induced by stacking faults in the InGaN layer due to strain relaxation[5].

In the present work, graded InGaN buffers are employed to relax the strain arising from the lattice and thermal mismatches between GaN/InGaN epilayers grown on sapphire. The primary objective of this study is to investigate the influence of graded InGaN on the strain and the shift of the photoluminescence (PL) spectra of a GaN epilayer grown on top of it. Using micro-Raman spectroscopy, the top GaN layer of the structures with different InGaN grading were found to be under compressive strain. However, this strain was much lower compared to the conventional GaN thin films grown on sapphire with a low-temperature GaN or AlN buffer. The PL results further confirmed the compressive nature of the GaN cap layer.

2. EXPERIMENT

The samples were grown by metalorganic chemical vapor deposition (MOCVD) on *c*-plane sapphire substrates with a 25 nm thick low temperature GaN nucleation layer. During the growth, the reactor pressure was kept at 200 Torr. A high temperature GaN layer of thickness 500 nm was grown on top of the nucleation layer before the graded InGaN layers were grown. Structure I has five layers, each 200 nm thick $In_xGa_{1-x}N$ and separated from one another by a thin 10 nm GaN grown at 750 ^{0}C and annealed at 1000 ^{0}C. This thin layer shall be termed as the insertion layer. Structures I and II were capped with a 100 nm GaN grown at 1000 ^{0}C while the GaN cap layer of Structure IV was grown at a condition similar to the insertion layer. A control structure consists of only pure GaN with a thickness of 800 nm, was also grown for a comparative analysis. The information for some of the InGaN layers as well as the GaN caps is shown in Table I.

Table I. Growth parameters and data from SEM, micro-Raman spectroscopy and PL in all the structures (RT stands for room temperature).

Structure	I	II	III	IV	Control structure
InGaN layer thickness (nm)	200	200	100	200	Nil
GaN cap layer thickness (nm)	100	100	Nil	10	Nil
GaN cap layer growth temperature (^{0}C)	1000	1000	Nil	750	Nil
V-pit density (cm^{-2})	0	2.5 x 10^7	5 x 10^9	1 x 10^6	Nil
E_2(TO) (cm^{-1})	567.6	567.9	-	-	568.0
ε_{xx} (x 10^{-4}) ε_{zz} (x 10^{-4})	–0.49 +0.26	–3.24 +1.72	- -	- -	–3.91 +2.08
InGaN RT PL peak energy (eV)	3.267	3.289	-	-	NA
GaN RT PL peak energy (eV)	3.410	3.421	-	-	3.427

3. RESULTS AND DISCUSSIONS

Figure 1 illustrates the E_2(TO) phonons from GaN for Structure I, II and the control structure under the $z(xx)\bar{z}$ scattering configurations excited by a 514.5 nm laser line. The z-axis is parallel to the crystal c-axis. From symmetry consideration, for uniaxial strain along the c-axis or biaxial strain in the plane normal to the c-axis, $\varepsilon_{xx} = \varepsilon_{yy} = \varepsilon_{zz}$ and $\varepsilon_{xy} = \varepsilon_{yz} = \varepsilon_{xz} = 0$, where ε_{ij} denotes the strain tensor components. ε_{xx} and ε_{zz} are the in-plane strain and normal strain, respectively. The strain can be estimated using the equation, $\varepsilon_{xx} = \Delta\omega_\lambda /(2p_\lambda + q_\lambda r)$ where $\varepsilon_{zz} = r\varepsilon_{xx}$ and $r = -2C_{13}/C_{33}$.[5] $\Delta\omega_\lambda$ is the observed shift of the E_2(TO) phonon mode while p_λ and q_λ are the deformation potential constants. The reference strain-free frequency for the E_2(TO) phonon mode is 567.5 cm^{-1} (400 μm HVPE GaN bulk epilayer, IMRE standard), shown as the vertical dashed line in Fig. 2(a). The following values were used for the calculations: C_{13} = 106 GPa, C_{12} = 145 GPa and C_{33} = 398 GPa[6] while p_λ = –812 ± 25 cm^{-1} and q_λ = –920 ± 60 cm^{-1} per unit strain at room temperature.[7] The in-plane and normal strain components are shown in Table I. All the three structures have a compressive GaN in-plane residual strain component. The residual strain in the GaN of Structure I is much smaller than that of Structure II and the control structure; therefore, strain relaxation is most effective in Structure I. The function of the insertion layer in Structure I is to steadily relax the strain and annihilate the dislocations that build up in each 200 nm thick InGaN layer. With a more gradual grading rate of the InGaN buffer in Structure I (1% In/210 nm) than in Structure II (5% In/200 nm), the strain relaxation is therefore more effective. If the grading is too rapid as in Structure II, the number of dislocations in the InGaN increases more quickly, thus increasing the probability of the dislocations becoming entangled and hindering the motion of the dislocations to relax the strain in the InGaN layer.

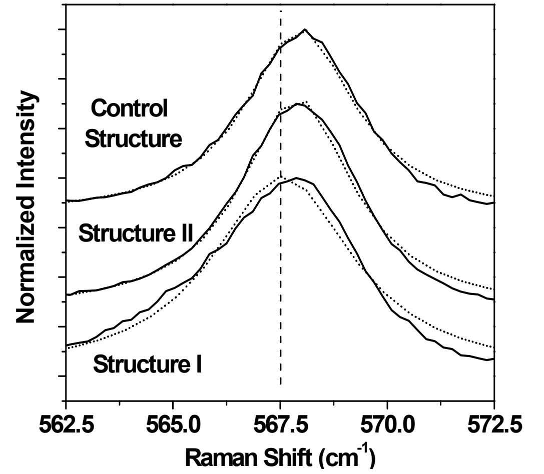
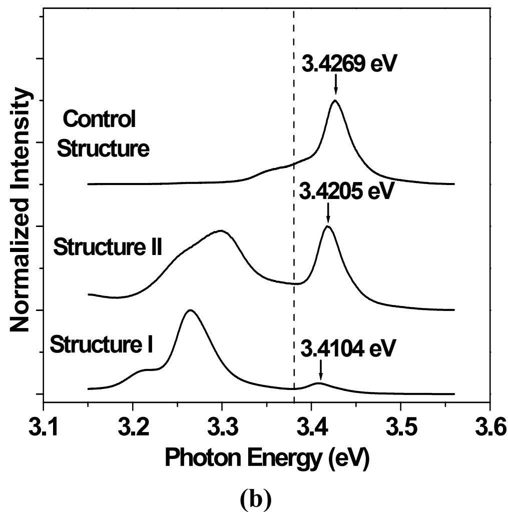


Figure 1: Strain relaxation in Structure I, II and control structure is illustrated with shifts in Raman spectra of the GaN E_2(TO) phonon . The dashed line is the strain-free reference E_2(TO) phonon mode at 567.5 cm^{-1} (IMRE standard) for a 400 μm HVPE GaN bulk epilayer. The dotted line represents fitting of the spectra taking into consideration of the slit width and spatial correlation.

To confirm the results obtained by Raman scattering in off-resonant conditions, the strain state was also investigated by UV-PL measurements. A very direct method to determine qualitatively the strain state of the GaN and InGaN is to measure their band edge PL energy, which should increase with the compressive strain. PL was excited with a 325 nm line of a 10 mW He-Cd laser. Figure 2(b) illustrates the room temperature PL spectra for Structure I, II and the control structure. The dashed line shows the PL line (3.39 eV) for a completely relaxed GaN bulk layer at room temperature.[8-10] Two main emission lines were observed. The lower-energy line is due to the PL from the $In_xGa_{1-x}N$ layer while the higher-energy line is due to the PL from the GaN layer. The GaN PL peaks in our samples show a blue shift with respect to that of the completely relaxed bulk GaN layer; hence confirming the compressive strain characteristics in the GaN cap layers. However, the GaN peaks of Structure I and Structure II show red shift with respect to that of the control structure. Structure I shows a larger amount of red shift, substantiating the result obtained by micro-Raman spectroscopy indicating that Structure I produces a greater amount of strain relaxation in the GaN cap layer than that of Structure II.



(b)

Figure 2: Room temperature Photoluminescence spectra of structure I, II, and control structure. The dashed line is the PL line (3.39 eV) for a completely relaxed GaN bulk layer at room temperature.[8-10] Peak positions were estimated from the multiple-curve fitting.

Figure 3 depicts the Scanning Electron Microscopy (SEM) pictures of the structures, showing the V-pits on the surfaces. The V-pit densities of all the structures are shown in Table I. The control structure does not have any V-pits but Structure III does; hence verifying that the V-pits indeed originate from the InGaN layer. Structure I showed no observable surface V-pits while the V-pit density in Structure IV is three orders of magnitude lower than that of Structure II. Growing the 200 nm InGaN and 10 nm GaN layers in Structure I at a slower In grading rate and with an annealing step carried out after the insertion layer, results in a structure which annihilates more dislocations. As V-pits originate from threading dislocations, they are effectively eliminated. The only difference between Structure II and IV is the thickness and growth conditions of the GaN cap layer. Since the GaN cap layer in Structure IV is the insertion layer, the significant reduction of V-pit density in Structure IV as compared to Structure II implies that the growth conditions of the insertion layer play an important role in suppressing and filling the V-pits in the GaN cap layer. The optimum growth conditions were empirically found to be a thin 10 nm GaN grown at 750 ^{0}C and annealed at 1000 ^{0}C. With more insertion layers grown, more dislocations and V-pits were annihilated and filled until a point whereby no more V-pits were observed on the surface, as seen in Structure I.

4. CONCLUSION

Graded InGaN epilayers present an enhanced performance in relaxing the strain in the InGaN/GaN epilayers grown on sapphire, as compared to conventionally grown GaN. The growth technique employed in Structure I also enhances the luminescence intensity from the InGaN layers. A subsequent decrease in the room temperature PL linewidth was clearly observed in Figure 2 of Structure I. The intensity enhancement of the PL from InGaN and the observed linewidth reduction suggest improvement in the crystalline quality apart from the strain relaxation. Such a graded buffer growth technique would be useful to grow high quality strain released GaN epilayers for optoelectronic devices.

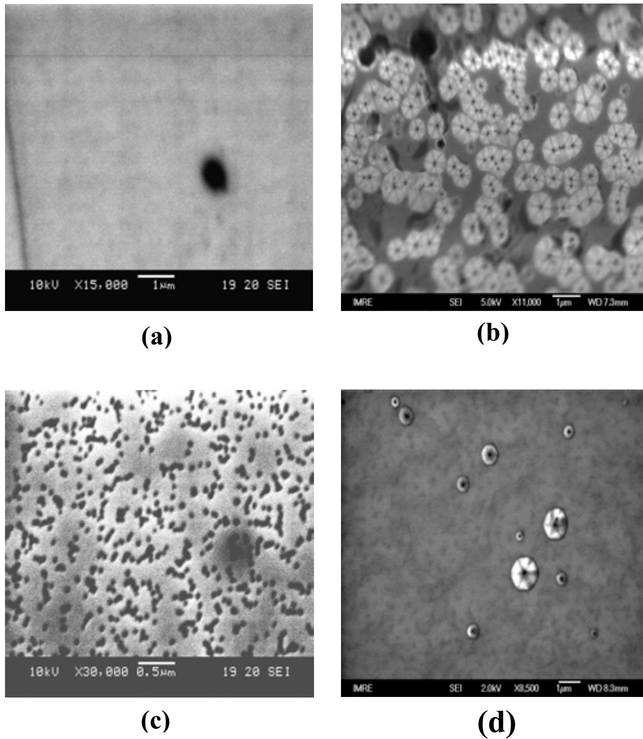


Figure 3: SEM pictures showing the surface V-pits of:
(a) Structure I, (b) Structure II,
(c) Structure III, and (d) Structure IV.

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