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

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ABSTRACT

Graded InGaN buffers are employed to relax the strain arising from the lattice and thermal mismatches between GaN/InGaN epilayers grown on sapphire. The formation of V-pits in linearly graded InGaN/GaN bulk epilayers is illustrated. The V-pits were sampled using Atomic Force Microscopy and Scanning Electron Microscopy to examine their variation from the theoretical geometry shape. We discovered that the size of the V-pit opening in linearly graded InGaN, with and without GaN cap layer, has a Gaussian distribution. As such, we deduce that the V-pits are produced at different rates, as the growth of the InGaN layer progresses. In Stage I, the V-pits form at a slow rate at the beginning and then accelerate in Stage II when a critical thickness is reached before decelerating in Stage III after arriving at a mean size. It is possible to fill the V-pits by growing a GaN cap layer. It turns out that the filling of the V-pits is more effective at lower growth temperature of the GaN cap layer and the size of the V-pits opening, which is continued in to GaN cap layer, is not dependent on the GaN cap layer thickness. Furthermore, graded InGaN/GaN layers display better strain relaxation as compared to conventionally grown bulk GaN. By employing a specially design configuration, the V-pits can be eliminated from the InGaN epilayer.

1. INTRODUCTION

GaN and InGaN are wide band gap semiconductor materials suitable for the whole range of visible-light and ultraviolet optoelectronic devices. Bulk GaN films are known to contain a high density of defects primarily threading dislocations, due to the large lattice mismatch and thermal expansion coefficient difference between the epilayer and the substrate. These defects affect both electrical and optical properties of the material, for instance, broken bonds at defect sites may enhance free carrier recombination. A low temperature grown AlN or GaN buffer layer considerably improves the crystal quality of the GaN epilayers but the films still contain a threading dislocation (TD) density of about 10^8 – 10^{10} cm^{-2} [1]. For InGaN, the most regularly observed defects 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 [2]. 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 [2]. a shall be referred to as the radius of the hexagon hereafter. V-pits are believed to have originated from either TD [3] or generated from the stacking mismatch boundaries induced by stacking faults in the InGaN layer due to strain relaxation [4].

In the present work here, graded InGaN buffers are employed to relax the strain arising from the lattice and thermal mismatches between GaN/InGaN epilayers grown on sapphire. We report on the formation of V-pits in linearly graded bulk InGaN/GaN layers, based on the statistics of the size distribution of the V-pit openings obtained from Atomic Force Microscopy (AFM) and Scanning Electron Microscopy (SEM). Using MicroRaman Spectroscopy, we can also observe that the InGaN and GaN layers exhibit greater strain relaxation than in the conventional high temperature GaN.

2. EXPERIMENT

The samples were grown by Metalorganic Chemical Vapor Deposition (MOCVD) on *c*-plane sapphire substrates with a 25 nm low temperature GaN nucleation layer. The reactor pressure was 200 Torr. A high temperature GaN layer of thickness 500 nm is grown atop the nucleation layer before the linearly graded InGaN layer is grown. All InGaN epilayers are graded from 0 %In and grown at 750 ^{0}C. Depending on circumstances and objectives, a GaN cap layer is deposited at different temperatures.

The samples grown for the V-pit study have parameters shown in Table I while Table II shows the strain relaxation comparison of the samples. A Control structure (not listed in Table I), consists of only pure GaN with a thickness of 800 nm, is also grown. Structure IV is described in Section 3B.

Table I: Growth parameters as well as the AFM and SEM statistics for the $In_xGa_{1-x}N$ layers and the GaN caps. This is for the purpose of V-pits sampling. All the InGaN layers are linearly graded from $x = 0$ to the desired final In composition.

Sample		I	II	III
InGaN Linear Grading Rate (% In/μm)		25	25	25
Thickness (nm)		200	100	200
GaN cap layer	Thickness (nm)	100	Nil	10
	Growth Temperature (^{0}C)	1000	Nil	750
V-pit density (cm^{-2})		2.5×10^7	5×10^9	1×10^6
h/a		0.2176	0.2749	0.2086
Mean size of a (nm)		93.885	45.536	91.498

Table II: Estimated strain tensor components from Raman shift in E_2(TO) for strain relaxation comparison. Structure IV is described in section 3B

Sample		e_{xx} (x 10^{-4})	e_{zz} (x 10^{-4})
I	V-pits	1.736 (compressive)	0.394 (tensile)
	Bulk	3.223 (compressive)	1.717 (tensile)
IV		0.496 (compressive)	0.264 (tensile)
Control		3.884 (compressive)	2.069 (tensile)

3. RESULTS AND DISCUSSIONS

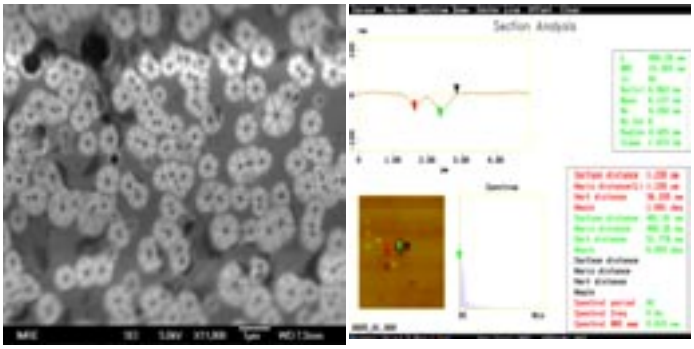
A. V-PITS FORMATION AND FILLING

The formation of V-pits was investigated using AFM and SEM. All structures showed observable V-pits, except the control structure. This verified that the V-pits indeed originate from the linearly graded InGaN bulk layer. The location of the V-pits is randomly distributed; some surface areas tend to have more pits while some have even and relatively smooth surfaces. The V-pits formed in all the structures violate the theoretical geometry shape [2]. Fig. 1 shows the SEM and AFM pictures of some of the samples. For a start, not all V-pits appear symmetrically hexagonal. Therefore, for the purpose of sampling and calculation, the only V-pits which are hexagonal are considered.

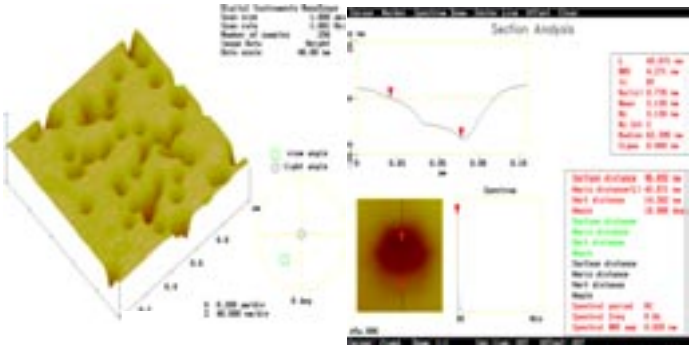
The V-pits were sampled to investigate the deviation from the theoretical geometry shape. The radius, *a* of the V-pits for all the structures, demonstrates a Gaussian distribution. The mean value of *a* vary with the final Indium composition and the total thickness of the linearly graded InGaN bulk layer, but not the GaN thickness, as specified in Table I. The radius of the V-pits approximately double when the thickness of the linearly graded InGaN is doubled. With a Gaussian radii distribution, we can deduce a mechanism for the formation of V-pits in linearly graded InGaN epilayers. We propose that the V-pits are generated at different rates, as the MOCVD growth of InGaN proceeds, as represented in Fig. 2. In Stage I, the V-pits form at a low rate at the beginning and then accelerate in Stage II when a critical thickness is reached before decelerating in Stage III after arriving at a mean size. The opening of the V-pits increases in dimension as the epilayer thickness increases. This means that V-pits with radii considerably greater than the mean values are initiated from lower layers, and vice versa.

The lattice mismatch between InGaN and the underlying GaN epitaxial layer has an immense impact on this effect. Indium atoms tend to phase-segregate to the six $\{10\bar{1}1\}$ planes to reduce the deformation energy [5] due to the lattice mismatch during the InGaN growth. V-pits with radii substantially larger than the mean values are very infrequent because the initial lattice mismatch between the InGaN graded layer and the underlying GaN layer is very insignificant. As such, the InGaN lattice prefers to contain the strain energy due to lattice mismatch until a critical thickness when it is favorable to release the stress by forming V-pits. Defect formation accommodates the deformation energy stored in the InGaN layer owing to the lattice mismatch, and hence reduces the composition pulling effect [6]. As a new layer of InGaN with a different In composition is deposited, the preceding V-pits will extend in a V-shaped manner, while more new V-pits are nucleated to relax more strain. Immediately upon reaching the critical thickness, V-pits nucleation will accelerate extensively because the system intends to release the strain within the graded InGaN layer at the fastest rate possible because V-pits formation is the most efficient mode. This is the reason for observing a rapid climb in the number of V-pits until the mean radii value is reached where V-pits formation will start to diminish after sufficiently relaxing the graded InGaN. One of the probable reasons for the V-pits to be distorted is because adjacent V-pits combine into one. This process sustains until a stage when the strain produced due to the lattice mismatch is completely relaxed. This is the primary reason for observing only very few V-pits with radii significantly smaller than the mean values. With a same grading rate, as the In final composition and the thickness of the InGaN layer increases, the mean value of *a* will shift to a larger value, as shown in Table I and Figure 2(b) (compare Sample I and Sample II, or Sample II and Sample III), because more deformation energy needs to be relieved.

The GaN cap layer is capable of filling up the V-pits at a random rate. Without a GaN cap layer, Structure II has a V-pit density of $5\text{x}10^9$ cm^{-2}, almost comparable to the typical threading dislocation density in nitride epilayers. With a GaN top layer, the density of the V-pit is reduced appreciably – the V-pit density is $2.5\text{x}10^7$ cm^{-2} in Sample II and $1\text{x}10^6$ cm^{-2} in Sample III. As the GaN layer starts to grow, most pits are "filled" randomly. Since the final hexagonal pits' shape is "partially deformed", it can be concluded that the (0001) surface grows at a faster rate than the $(10\bar{1}1)$ facets. Consequently, as the GaN epilayer growth progresses, the ratio, *h*/*a* of the V-pits decreases and distort the initial shape, as illustrated in Fig. 2(a). At the growth temperature of 1000 ^{0}C in Sample I, the Ga and N atoms may be too energetic, and as a result, the probability of filling the pits are more random. By lowering the usual growth temperature to 750 ^{0}C, the V-pit density in Structure III is reduced significantly. The effects of V-pit formation with growth temperature will be reported in a future publication.



(a)



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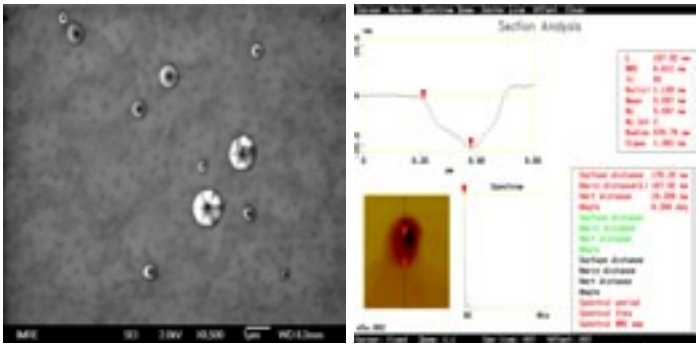
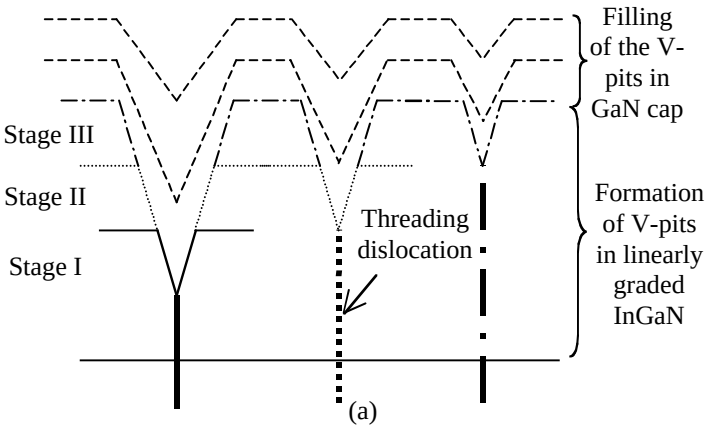
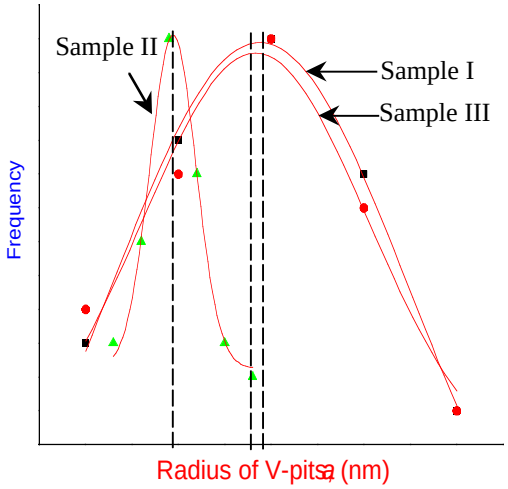


(c)

Fig. 1: AFM and SEM pictures of
(a) Sample I (b) Sample II and (c) Sample III



(a)



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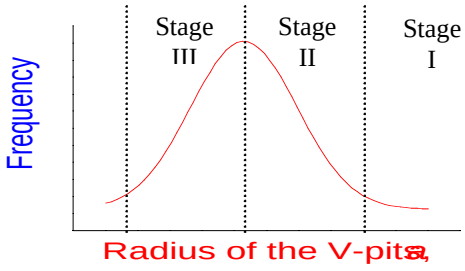

(c)

Fig. 2: A model for V-pits generation and filling:
(a) *Formation of V-pits in linearly graded InGaN layer*, showing Stage I, Stage II and Stage III.
Filling of V-pits in the GaN cap layer: The initial shape of the V-pits is distorted due to the filling mechanism of the GaN cap layer.
(b) The Gaussian distributions for the V-pits of Sample I, Sample II and Sample III. The long-dashed lines are the mean values the respective samples, as shown in Table I.
(c) A typical Gaussian distribution of the V-pits formation showing Stage I, Stage II and Stage III, respectively.

B. STRAIN RELAXATION

Structure IV has 5 layers of $In_xGa_{1-x}N$, with each layer separated by a layer of GaN grown at 750 ^{0}C. The first InGaN layer has x = 0.01, the second layer x = 0.02 and so on. A GaN cap is grown atop the final InGaN layer.

Structure IV was designed with an initial purpose to prevent tensile crack in a normal conventional GaN layer. As the film thickens during growth, the elastic strain energy in the film increases until, once a critical thickness is exceeded, it becomes energetically favourable to introduce misfit dislocations at the interface between the film and the substrate. When the lattice mismatch is large, as it is in the GaN/sapphire system (approximately 14% mismatch), it is energetically favourable for the film to grow by the nucleation of individual and isolated islands rather than as a continuous film. This is because the critical thickness is exceedingly small. Indeed, measurements indicate that the critical thickness for a coherent GaN film on sapphire is exceeded in growing a single monolayer. As the film continues to grow, the individual islands grow both in thickness and laterally until they coalesce.

Light scattering technique is an efficient method to investigate the vibration properties of III-V nitride semiconductors. Raman scattering is known to be a sensitive technique, in comparison to photoluminescence, for determining strain in epitaxial layers. The epitaxial layer in each structure, grown on (0001) sapphire substrates have residual strain resulting from the lattice and thermal mismatch between the substrate and the epilayer. The presence of elastic strain within the layers can give rise to shift and spreading of the phonon modes due to a change of interaction between the elastic medium and the macroscopic elastic field [7]. From symmetry consideration, for uniaxial strain along the hexagonal axis or biaxial strain in the plane normal to the hexagonal axis, $e_{xy} = e_{yz} = e_{xz} = 0$, where e_{ij} denotes the strain tensor components.

E_2(TO) phonon mode is applied here as reference to calculate the strain of the GaN cap layer. E_2 mode is employed because it produces the clearest signal as compared to other modes. LO phonons are not used because they vary with different impurity concentration. In this case, even though the layer is undoped, there will always be a residual oxygen concentration in the epilayer. Higher oxygen concentration in the layer produces a large amount of oxygen related point defects in the sapphire top monolayers. Therefore, during growth, oxygen may diffuse from sapphire into the buffer and further into the GaN layer. Higher defect concentration makes it easier for oxygen to diffuse within the buffer and the GaN layer. Less oxygen is available at the nitridated sapphire surface in the case of low temperature growth. Oxygen may lead to the formation of cubic GaN complexes within the hexagonal GaN, due to a high concentration of oxygen impurities.

The strain can be estimated using the equations from Davydov et al. [7] with reference to the 400 μm HVPE GaN bulk epilayer (IMRE standard), where the strain-free frequency for E_2(TO) phonon mode = 567.5 cm^{-1}. The following values are used for this prediction: C_{13} = 106 GPa and C_{33} = 398 GPa [8]; while p_λ = -850 ± 25 cm^{-1} and q_λ = -920 ± 60 cm^{-1} at room temperature [7]. The in-plane and normal strain components are shown in Table II. Referring to Table II, the strain in each structure is virtually negligible. The compositional disorder is therefore very small due to the unappreciable Raman-shift. However, it can be deduced that graded layers demonstrate a better strain relaxation as compared to the conventionally grown GaN (Control structure). Strain relaxation is most effective in Sample IV. For Sample I, it can be observed that the hexagonal V-pit area is more relaxed as compared to the as-grown GaN epilayer because it experiences lesser right Raman-shift. Another interesting aspect worth mentioning is that Sample IV does not contain any distinct V-pits. This means that the design as employed in Sample IV is capable of eliminating V-pits. The reason behind this phenomenon is still under investigation.

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. By compiling the statistics of the V-pit sizes observed in the linearly graded InGaN layers, the formation of V-pits is illustrated. It turns up that V-pits can be eliminated from the InGaN epilayer by employing a specially designed configuration, although this phenomenon is still under investigation.

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