

High Indium Concentration InGaN/GaN Grown on Sapphire Substrate by MOCVD

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Abstract Scanning electron microscopy, room temperature photoluminescence, and X-ray diffraction techniques have been used to characterize InGaN layer grown on InN and InGaN buffers. The growth was done on c-plane sapphire by MOCVD. Results showed that green emission was obtained which indicates a relatively high In incorporation.

Index terms high In $\text{In}_x\text{Ga}_{1-x}\text{N}$, highly-mismatched systems, strain relaxation, V-pits.

I. Introduction

The InGaN system provides the opportunity to fabricate light emitting devices over the whole visible and ultraviolet spectrum due to band-gap energies E_g between 3.42 eV for GaN and 1.89 eV for InN [1]. However, the fabrication of laser diodes was up to now restricted to the green, blue, and UV range although the emission toward the red region would be highly desirable, which can only be achieved with high In incorporation. This is partially related to the fact that the relationship between structural properties, In distribution, and light emission is still not completely clarified, and high In content in InGaN layers will result in a significant degradation of the crystalline quality of the

epitaxial layers. In addition, unlike other III-V compound semiconductors, the ratio of gallium to indium incorporated in InGaN is in general not a simple function of the metal atomic flux ratio, $f_{\text{Ga}}/f_{\text{In}}$. Instead, In incorporation is complicated by the tendency of gallium to incorporate preferentially and excess In to form metallic droplets on the growth surface [2]. This phenomenon can definitely affect the In distribution in the InGaN system.

From the previous work [3], it was found out that the formation of V-pits has been identified as the dominant strain relief mechanism in InGaN instead of the generation of misfit dislocation or even the motion of dislocation. V-pits formation will always take place in any InGaN film with a reasonable lattice mismatch; because of the existence of strong driving force at a critical point upon surpassing a certain thermal barrier. The system is energetically favorable to nucleate V-pits until a saturation point is reached.

This problem, however, motivates further research on a method to initially relieve a large amount of lattice mismatch in the $\text{In}_x\text{Ga}_{1-x}\text{N}$ without using dislocation glide. This method must also minimize V-pits formation as it will destroy the surface morphology if strain relief occurs over a large range of thickness. The primary objective of this research work is to obtain a

thick, planar, and relaxed $\text{In}_x\text{Ga}_{1-x}\text{N}$ epilayer with high In composition that can operate at longer wavelengths, particularly the red-color emission.

II. Experimental

All the samples were grown with different thicknesses of $\text{In}_x\text{Ga}_{1-x}\text{N}$ and composition of In by metalorganic chemical vapor deposition (MOCVD) on c-plane (0001) sapphire. Reduced threading dislocation (TD) densities of GaN was obtained by growing first a thin amorphous layer of GaN on top of the sapphire substrate to relax and confine defects generated by the large mismatch between the GaN and the sapphire. Then the subsequent growth was to grow the various $\text{In}_x\text{Ga}_{1-x}\text{N}$ layers.

III. Results and discussion

The first sample contained a thin InN or high In composition $\text{In}_x\text{Ga}_{1-x}\text{N}$ nucleation layer grown at low temperature on top of the high temperature GaN to create an amorphous layer (Fig. 1.a). This amorphous layer was expected to function like the low temperature grown GaN buffer layer, to contain all the defects and relax the misfit strain due to lattice mismatch. The subsequent growth should not nucleate V-pits if this initial layer has reduced the lattice-mismatch sufficiently so that strain in the $\text{In}_x\text{Ga}_{1-x}\text{N}$ film is below the critical strain needed for V-pit formation. The desired $\text{In}_x\text{Ga}_{1-x}\text{N}$ epilayer can be subsequently grown thereafter, lattice-matched to the thin InN or $\text{In}_x\text{Ga}_{1-x}\text{N}$ buffer. However, the first effort to grow the amorphous InN led to the formation of In droplets.

Sample II (Fig. 1.b) was then grown to follow up the first sample. A 40-nm $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}$ layer was grown at 700 °C before

growing the amorphous InN because of the difficulty in growing InN directly on top of the GaN. This lower In composition InGaN layer was expected to help in the nucleation.

| | |
|------------------------------|--|
| InN 560C | InN 560 °C |
| GaN 1000 °C, 1 μm | $\text{In}_{0.20}\text{Ga}_{0.80}\text{N}$, 700 °C, 40 nm |
| GaN, 530 °C, 25 nm | GaN 1000 °C, 1 μm |
| Sapphire | GaN, 530 °C, 25 nm |
| (a) | Sapphire |
| | (b) |

Fig. 1. Structure and growth condition of (a) Sample I and (b) Sample II.

As clearly shown in the XRD (Fig. 2) and SEM (Fig. 3.a) images, In droplets were still formed on the surface. Based on Ting et al. [4], adequate ammonia pressure must be maintained to prevent In surface segregation and the formation of droplets since the equilibrium nitrogen vapor pressure for InN is higher than that for GaN. This explanation is also supported by Ou et al., who argued that the relatively weak InN bond may also play a role in In surface segregation and, in the extreme case, insufficient ammonia can lead to the formation of droplets [5-6].

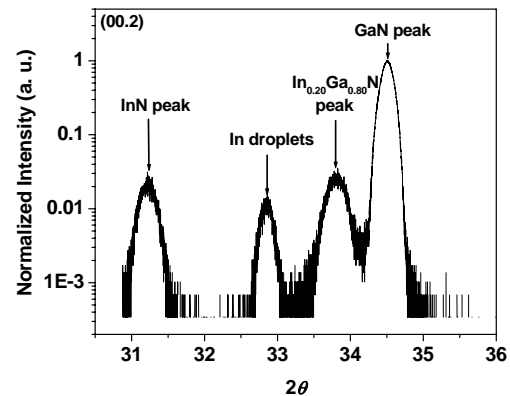
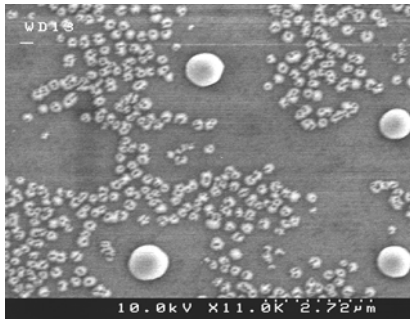
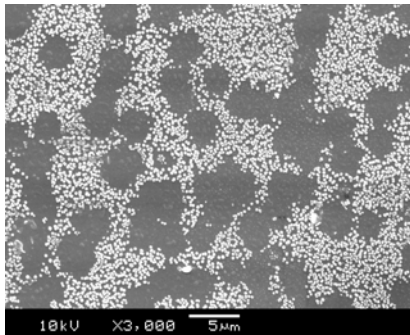


Fig. 2. The X-Ray Diffraction of Sample II.

This In-droplets can actually be etched away using dilute HCl for about 20 minutes, as shown in (Fig. 3.b).



(a)



(b)

Fig. 3. The SEM image of Sample II with (a) before and (b) after etching with HCl.

Sample III is similar to Sample II, but with a 200 nm-thick-InGaN layer grown at 750 °C on top of the InN layer (Fig. 4).

| |
|--|
| In _x Ga _{1-x} N, 750 °C, 200 nm |
| Ga flux 6 sccm |
| InN 560 °C, 20-30 nm |
| In _{0.20} Ga _{0.80} N, 700 °C, 40 nm |
| GaN 1000 °C, 1 μm |
| GaN, 530 °C, 25 nm |
| Sapphire |

Fig. 4. Structure and growth condition of Sample III.

From the SEM image (Fig. 5), it can be seen that the surface was full of V-pits, while the In-droplets disappeared, and have probably merged with the top InGaN layer. This was confirmed by the XRD as only 2 peaks were appeared (Fig. 6).

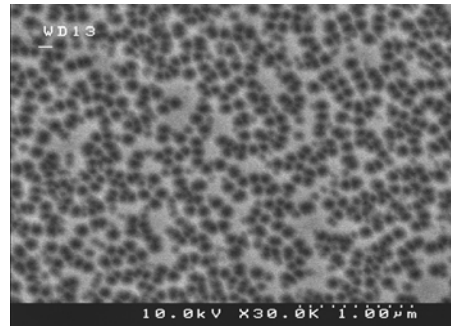


Fig. 5. The SEM image of Sample III showing the high density of V-pits.

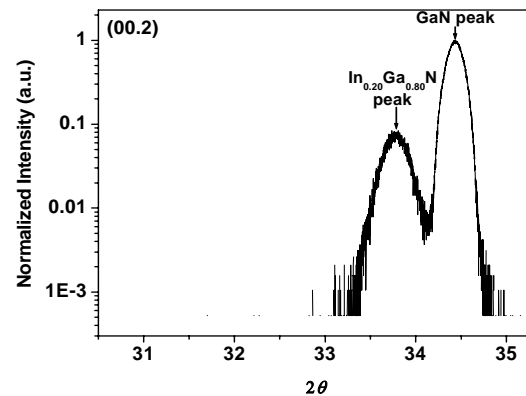


Fig. 6. The X-Ray Diffraction of Sample III.

RT-Photoluminescence technique was also used to compare the emission of Sample II and III. Seen from Fig. 7 below, green emission was obtained for Sample III, which indicates a relatively high In incorporation.

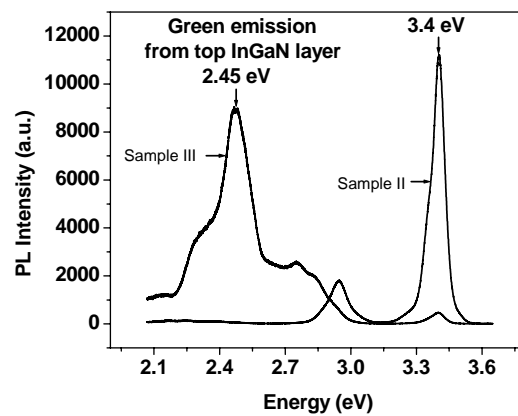


Fig. 7. RT-Photoluminescence spectra of Sample II and III.

IV. Conclusion

Thin amorphous-InN layer was employed to increase the In concentration in the InGaN layer, with a thin InGaN buffer grown before the InN. This resulted in green emission as shown by the RT-Photoluminescence spectra of Sample III.

Reference

1. D. Gerthsen, B. Neubauer, A. Rosenauer, T. Stephan, H. Kalt, O. Schon, and M. Heuken, *Appl. Phys. Lett.*, 79, 2552 (2001).
2. D. F. Storm, C. Adelman, and B. Daudin, *Appl. Phys. Lett.*, 79, 1614 (2001).
3. T. L. Song, S. J. Chua, E. A. Fitzgerald, P. Chen, and S. Tripathy, *J. Vac. Sci. Technol. A* 22(2), 287 (2004).
4. S. M. Ting, J. C. Ramer, D. I. Florescu, V. N. Merai, B. E. Albert, A. Parekh, D. S. Lee, D. Lu, D. V. Christini, L. Liu, and E. A. Armour, *J. of Appl. Phys.*, 94, 1461 (2003).
5. J. Ou, W. K. Chen, H. C. Lin, Y. C. Pan, and M. C. Lee, *Jpn. J. Appl. Phys., Part 2*, 37, L633 (1998).
6. S. M. Bedair, F. G. McIntosh, J. C. Roberts, E. L. Piner, K. S. Boutros, and N. A. El-Masry, *J. of Crystal Growth* 178, 32-44 (1997).