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Spatial control of InGaN luminescence by MOCVD selective epitaxy

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Abstract

Selective epitaxial growth of InGaN alloys was performed by metalorganic chemical vapor deposition (MOCVD). Templates consisted of arrays of circular etched holes in a SiO₂ mask layer, with pre-grown GaN hexagonal pyramid structures. The room temperature (300 K) photoluminescence (PL) peak wavelength increased with increasing mask-opening spacing for a constant mask-opening diameter. For 5 μ m diameter pyramids, peak PL wavelengths ranged from 457 to 505 nm for mask opening spacings of 6 to 13 μ m, compared to PL peak wavelength of 420 nm for a planar reference sample. © 1998 Elsevier Science B.V. All rights reserved.

1. Introduction

The application of selective epitaxy to the fabrication of electronic and optoelectronic devices is well established. The benefits of selective epitaxy have been realized for a variety of materials systems. Selective epitaxy of InGaAs alloys results in alloy compositions that are directly determined by the geometry of the mask [1]. This phenomenon allows the fabrication of complex three-dimensional optoelectronic devices.

Selective epitaxy of GaN was first reported in 1994 [2], and is becoming increasingly important for fabrication of devices. We have previously reported on the first demonstration of GaN selective epitaxy for electron field emission devices [3,4], and the fabrication of regrown ohmic contacts for fields-effect transistors (FETs) [5]. While selective epitaxy of nitride materials has been proposed for fabrication of optoelectronic devices [6], and optical properties of selectively grown GaN have been reported [7], no report yet exists on the selective epitaxy of InGaN. In this paper we will report for the first time on the spatial control of In incorporation in InGaN through the use of selective epitaxy. These results will not only benefit the development of GaN-based optoelectronic devices, but may also enhance the understanding of InGaN epitaxial growth in general.

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2. Procedure

Base GaN films for these studies were obtained using atmospheric pressure metalorganic chemical vapor deposition (MOCVD). The growth conditions for these films have been published elsewhere [8]. Plasma-enhanced chemical vapor deposition (PECVD) or electron beam evaporation was then used to deposit 200 nm thick SiO₂ films on GaN. These films were patterned using standard photolithographic procedures and etching with hydrofluoric acid (HF). A short (~ 8 s) dip in dilute HF was used immediately prior to regrowth in order to clean the growth surface. The selective epitaxy mask used for this study consisted of circular mask opening arrays, each in a close-packed configuration. The mask opening diameters ranged from 2 to $12 \,\mu\text{m}$, and the spacings from 3 to $13 \,\mu\text{m}$. The array fill ratios, defined as the ratio of mask opening area to total array area, varied from 0.03 to 0.77. Selective epitaxy of GaN was performed by MOCVD using trimethyl gallium (TMGa) and ammonia (NH₃) precursors, and hydrogen as the carrier gas, at 76 Torr reactor pressure and at a growth temperature of 1000°C. A more detailed discussion of the growth conditions for GaN hexagonal pyramids is contained in Ref. [3]. The arrays of GaN pyramids were characterized using low-voltage field-emission scanning electron microscopy (FE-SEM).

Following solvent cleaning, and another short HF dip immediately prior to re-introduction into the MOCVD reactor, selective epitaxy of InGaN was performed using trimethyl indium (TMIn), TMGa, and NH₃ precursors. Growth was performed at atmospheric pressure and a growth temperature of 800°C. InGaN growth was immediately preceded by growth of a thin GaN layer at 76 torr and 1050°C. The resulting structures were characterized using FE-SEM and room temperature (300 K) photoluminescence (PL) spectroscopy. The PL excitation energy was 200 W/cm².

3. Results and discussion

We have previously reported on the growth anisotropies that occur during selective epitaxy of GaN by MOCVD. Hexagonal pyramid structures are the result of growth using circular mask openings with diameters of several microns. While the growth rate on the pyramid sidewalls is a function of growth conditions, this shape is consistently obtained. In all array regions, hexagonal pyramid structures were obtained following GaN growth. Under the conditions used for GaN pyramid array growth for this study, nuclei are absent from the mask surface for several hundred microns from the nearest exposed GaN surface. All pyramid array regions were thus free from mask nucleation following GaN growth.

Fig. 1 contains an SEM image of the pyramids following InGaN growth. The GaN pyramid morphology has been maintained during InGaN growth. The InGaN growth step was the equivalent of 100 nm of planar growth thickness, but no In-GaN growth rate calibration yet exists for pyramid sidewalls. While large masked regions were coated with InGaN nuclei ~ 10 μ m from the nearest exposed semiconductor surface, mask nuclei were absent from pyramid array regions except for those with fill ratio less than ~ 0.10.

Photoluminescence spectroscopy on the various arrays of GaN/InGaN pyramids revealed that, for a given mask-opening diameter, the peak luminescence wavelength was a function of the spacing between openings. A series of PL spectra obtained



Fig. 1. Scanning electron microscope (SEM) image of GaN/ InGaN pyramids (bar $=5\,\mu m).$

for arrays with 5 μ m diameter openings is shown in Fig. 2 for different spacings between openings. The peak intensities for all spectra were normalized for comparison. Fig. 3 contains a plot of the peak wavelength versus spacing for the arrays. The peak PL wavelength is a strong function of mask geometry, and luminescence from 457 to 505 nm can be controllably obtained from a single growth. For comparison, a planar growth reference sample exhibited peak emission at 420 nm.

The large differences in pyramid density in the different arrays also resulted in large differences in PL intensity. The peak PL intensity decreases with increasing spacing, with the most dense array exhibiting a peak intensity $\sim 5.8 \times$ that of the least dense array. The fill ratio of the most dense array is also $\sim 4.8 \times$ that of the least dense array. Since the InGaN film area is directly proportional to the fill ratio, this suggests that the real decrease in luminescence intensity over this ~ 50 nm wavelength range is not significant.

A detailed study of the optical properties of these GaN/InGaN pyramid arrays will be necessary to identify the recombination mechanisms responsible for the luminescence observed in Fig. 2. We believe, however, that the peaks occurring in the range from 450 to 500 nm are related to the InGaN band edge. These PL results would then be consistent with increasing incorporation of In in InGaN as the



Fig. 2. Photoluminescence (PL) spectra obtained from GaN/InGaN pyramid arrays with original mask opening diameters of $5 \mu m$, and various spacings between openings.



Fig. 3. Peak PL wavelength for GaN/InGaN pyramids obtained using $5 \,\mu m$ diameter mask openings and various spacings.

spacing between features is increased. Studies of the InGaAs system revealed that the In content of selectively grown structures increased with decreasing size and increasing spacing [9]. Thrush and co-workers attributed this effect to the greater tendency for complete decomposition of TMIn compared to TMGa. The trends we have discovered so far with the InGaN system correlate well with those observed for InGaAs.

The epitaxial growth of InGaN alloys with peak luminescence beyond 450 nm is highly challenging. This study has indicated that selective epitaxy of InGaN may be a useful technique for enhancing In incorporation, and thus increasing the emission wavelength of GaN-based optoelectronic devices. While this study has concentrated on InGaN growth on pyramid sidewalls, associated work has revealed that similar trends occur when selective epitaxy is performed with planar mask openings. The ability to spatially control InGaN composition will allow the development of more complex devices such as those which have been developed in the As- and P-based systems.

4. Conclusion

To summarize, selective epitaxial growth of In-GaN alloys has been performed by MOCVD. GaN pyramid shapes are unchanged following growth of thin InGaN layers. Perfect InGaN growth selectivity is achieved for regions with fill ratios > 0.10. Photoluminescence peak wavelength increased from 457 to 505 nm with increasing mask-opening spacing for a constant 5 µm mask-opening diameter. PL peak wavelengths of 420 nm were obtained for a planar reference sample, suggesting that increased In incorporation can result from selective epitaxy of InGaN. This demonstration of In incorporation enhancement and spatial control of emission wavelength will benefit the development of a next generation of non-planar optoelectronic devices from the (Al, Ga, In)N system.

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References

- [1] E.J. Thrush, M.A. Gibbon, J.P. Stagg, C.G. Cureton, C.J. Jones, R.E. Mallard, A.G. Norman, G.R. Booker, J. Crystal Growth 124 (1992) 249.
- [2] Y. Kato, S. Kitamura, K. Hiramatsu, N. Sawaki, J. Crystal Growth 144 (1994) 133.
- [3] D. Kapolnek, R.D. Underwood, B.P. Keller, S. Keller, S.P. DenBaars, U.K. Mishra, J. Crystal Growth 170 (1997) 340.
- [4] R.D. Underwood, D. Kapolnek, B.P. Keller, S. Keller, S.P. DenBaars, U.K. Mishra, Solid-State Electronics 41 (1997) 243.
- [5] Y.-F. Wu, D. Kapolnek, P. Kozodoy, S. Keller, B.P. Keller, S.P. DenBaars, U.K. Mishra, 24th Int. Symp. on Compound Semiconductors, San Diego, California, Sept. 1997.
- [6] T. Tanaka, K. Uchida, A. Watanabe, S. Minagawa, Appl. Phys. Lett. 68 (1996) 976.
- [7] X. Li, A.M. Jones, S.D. Roh, D.A. Turnbull, S.G. Bishop, J.J. Coleman, J. Electron. Mater. 26 (1997) 306.
- [8] B.P. Keller, S. Keller, D. Kapolnek, W.-N. Jiang, Y.-F. Wu, H. Masui, X.H. Wu, B. Heying, J.S. Speck, U.K. Mishra, S.P. DenBaars, J. Electron. Mater. 24 (1995) 1707.
- [9] J. Finders, J. Geurts, A. Kohl, M. Weyers, B. Opitz, O. Kayser, P. Balk, J. Crystal Growth 107 (1991) 151.