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Growth and Characterization of $In_xGa_{1-x}N$ MQW using a novel method of temperature gradient OMVPE

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Abstract

Composition-graded $In_xGa_{1-x}N$ Multiple Quantum Wells were deposited on single wafers using a novel growth technique called temperature gradient Organometallic Vapor Phase Epitaxy. A large temperature gradient, 710°C to 785°C, was imposed on 2" sapphire substrates while growing seven period $In_xGa_{1-x}N$ Multiple Quantum Well structures. Photoluminescence results show that high quality films, with variations in emission peak from 2.3 eV to 3.0 eV as a function of position, can be deposited on a single wafer. Photo-modulated transmission was used to determine band-edge optical transition as a means to obtain indium concentration as a function of position. Atomic Force Microscopy was used to image the morphology and relate indium concentrations to film microstructure and optical quality. This is a novel technique that uses a temperature gradient during Organometallic Vapor Phase Epitaxy for depositing variable emission wavelength devices on single wafers.

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Keywords: A1. Optical Spectroscopy; A1. Atomic Force Microscopy; A3. Metalorganic chemical vapor deposition; A3. Multiple quantum well; B1. Nitrides; B1. InGaN/GaN

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1. Introduction

Recently, there has been much progress in the fabrication and performance of optical devices based on Group III-Nitride technology [1,2]. Blue/green LEDs grown by Organometallic Vapor Phase Epitaxy (OMVPE) using InGaN/GaN Multiple Quantum Well (MQW) structures are already commercially available. Although these nitride based devices have been very successful, the continued progress of these LEDs is often limited by the lack of fundamental understanding of $In_xGa_{1-x}N$ deposition [3]. With the emergence of white LEDs, it is especially important that the fundamental issues for growing high quality $In_xGa_{1-x}N$ materials be understood [4,5].

Optical properties of LEDs are directly related to film quality which is easily influenced by the growth conditions and fabrication processes. OMVPE is the method of choice although it poses many challenging issues. InN decomposes at approximately 550°C under a flow of NH_3 and has a very low miscibility in GaN [6]. GaN is grown at temperatures in excess of 1000°C to obtain an acceptable cracking efficiency of ammonia. Compromise between these has led to growth temperatures for $In_xGa_{1-x}N$ ($0<x<1$) between 500°C - 800°C [7-9].

Aoyama and Sakai show that indium content and optical properties of InGaN structures can be altered by imposing temperature variations on the substrate [10]. But their technique requires physically altering the back of the substrate which is time consuming and financially detrimental in a production environment. In this study, a novel production method of temperature gradient OMVPE for $In_xGa_{1-x}N$ MQW structures was developed. High quality $In_xGa_{1-x}N$ MQW structures were deposited by imposing a large temperature gradient across the substrate while holding all other growth parameters constant. Optical and morphological characterization was performed as a function of position across the substrate in the direction of the temperature gradient. As a result, this technique allows deposition of variable emitting single wafer structures in the lateral direction. These novel devices have use for broadband wavelength emission/detection and solar cell technologies which can collect photons over a wide spectral range [11].

2. Experimental Procedure

$In_xGa_{1-x}N$/GaN MQW structures were grown on 2” (0001) sapphire substrates using a vertical flow EMCORE D180 MOCVD reactor. Trimethylgallium (TMG), Trimethylindium (TMI) and ammonia were used as the precursors for Ga, In, and N, respectively. N_2 was used as the carrier and dilution gases for the deposition of the active InGaN/GaN (30Å/150Å) regions to increase the amount of In incorporated while H_2 was used for all other layers. A low temperature GaN nucleation layer was deposited at 550°C followed by a ~2μm high temperature GaN intrinsic layer at 1050°C. This was followed by the growth of a 7 period InGaN/GaN MQW structure with a temperature gradient across the wafer from 710°C at the cool end to 785°C at the hot end. The structure is capped by the final barrier layer which is approximately 200Å of GaN. The gradient was imposed by adjusting the power input to two resistive heaters that are placed under the susceptor at opposite ends of the substrate position. The temperature at the end positions of the substrate were determined by IR pyrometry. Figure 1 shows a schematic of the substrate with temperature gradient. It is important to note that this reactor is capable of growing compositionally uniform $In_xGa_{1-x}N$ structures when a zero-temperature gradient is used.

Samples were analyzed using room temperature (RT) photoluminescence (PL) for emission output, RT photo-modulated transmission spectroscopy (PT) for band edge transition, and atomic force microscopy (AFM) for morphology characterization. The photoluminescence signals were generated by excitation with the 325 nm line of a HeCd laser (~ 3 mW). The signals were then dispersed by a 1 m double-grating monochromator and detected by a Hamamatsu R928 photomultiplier tube. For photo-modulated transmission spectroscopy experiments, quasi-monochromatic light from a xenon lamp dispersed by a 0.5m monochromator was focused on the surface of samples polished on the back side as a probing beam. An ultra-violet enhanced Si photo-diode was used as the detector while a chopped HeCd laser beam provided the modulation. AFM images were obtained using a Digital Instruments 3100 Scanning Probe Microscope with 160μm silicon tip cantilevers in Tapping Mode.

3. Results and Discussion

Figure 2 shows RT photoluminescence spectra for a sample taken at seven equally spaced regions across the 2” wafer. Region 1 corresponds to a growth temperature of 785°C and Region 7 corresponds to a growth temperature of 710°C where the regions in between are assumed to have a linear temperature change with position (See Figure 1). The data show that there is a systematic evolution of PL emission across the wafer due to the imposed temperature gradient. The high temperature end (Region 1) corresponds to the high energy emission. The peak PL emission energy decreases with decreasing growth temperature.

Figure 3 shows the peak emission energy as well as the PL intensity as a function of position. There is a much greater change in emission output for the high temperature range (Region 1-4) than for the low temperature range (Regions 5-7). It is also shown that the PL intensity is very high and relatively constant for this high temperature region while there is a large decrease in PL intensity as growth temperature decreases. This shows that the low temperature material has an apparent decrease in optical quality at a growth temperature below ~750°C. This implies that materials that must provide emission output at energies below ~2.5eV must have other growth conditions adjusted, such as Group-III molar flow rates, to compensate for decreased optical quality due to low temperature deposition.

To understand the change in material properties as a function of temperature, photo-modulated transmission was used. Figure 4 shows the PT data for each region across the wafer. The feature at high energy (~3.4 eV) is attributed to band-to-band transitions in the GaN barrier. The broad feature at lower energies correspond to band-to-band transitions taking place in the InGaN QW. As expected, there is a decrease in transition energy as a function of position. This is due to changes in indium concentration. Wu *et al.* [11] have determined a bowing parameter of 1.43 eV for the InGaN ternary alloy system. Using

$$Eg(x) = 3.42(1-x)+0.77x-1.43x(1-x) \qquad (1)$$

and neglecting quantum confinement and strain effects, indium concentration can be determined. Figure 5 shows the calculated indium concentration as a function of position across the wafer. It is shown that a film with $In_xGa_{1-x}N$ MQW structures with increasing In incorporation within a single wafer can be controllably deposited. More importantly, it implies that the growth temperature is the predominant parameter in determining In concentration, and therefore optical ouput, although more studies need to be performed regarding other growth parameters when combined with temperature gradient OMVPE.

AFM images were obtained to determine morphology for changes in growth temperature. Figure 6 shows AFM images (1μm x 1μm) for regions 1, 4, and 7 which correspond to the high temperature end, the mid-temperature, and the low temperature end of the wafer, respectively. Region 1 is smooth with an overall surface root-mean-square (RMS) roughness of only ~1.8 nm. Some small areas of inclusions commonly associated with dislocation propagation [12, 13] caused by lattice mismatch between the InGaN/GaN interfaces are observed. As the indium incorporation increases, due to decreased growth temperature, the roughness of the top surface of the cap layer increases as shown in region 4 and 7 with RMS surface roughness of 5.2 nm and 6.6 nm, respectively. This increase in surface roughness was consistent across the wafer for all seven of the regions tested. It appears that as more In is incorporated into the structure, the morphology of the area deteriorates resulting in a decrease in optical quality. Quantum well and barrier thicknesses for the different regions across the wafer are currently being investigated to determine the effect on optical properties.

4. Conclusion

A novel method of temperature gradient OMVPE has been developed for depositing high quality, compositionally graded $In_xGa_{1-x}N$ MQWs. A large temperature gradient was imposed across the substrate and was shown to vary In concentration in the lateral direction across the film. PL was used to characterize the output emission of the film as a function of position. It was shown that the output emission energy and the intensity decreased with decreasing temperature. PT was used to obtain In concentration as a function of position (temperature). As temperature decreases across the wafer, the In content increases. AFM was used to study the surface topology in the direction of the temperature gradient. It was shown that the surface roughness increases as the temperature decreases. This is due to the increasing In concentration gradient which increases the dislocation density due to the increased lattice mismatch between the $In_xGa_{1-x}N$ quantum well and the GaN barrier. This is also consistent with the decreases in optical quality as shown by the decreases in PL and PT intensities as In concentration increases. We have shown that temperature gradient OMVPE is a very promising technique for depositing variable emitting devices across a single wafer. Further studies for optimization of the optical properties at the higher In concentration (low T) are now being pursued.

Acknowledgement

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Figure Captions

Figure 1 – Schematic of temperature gradient imposed by varying input power to two electrical heaters located at the ends of the substrate within the vertical flow OMVPE reactor. Temperature gradient is assumed to be linear with Region 1 measured to be 785°C and Region 7 measured to be 710°C.

Figure 2 – Room temperature photoluminescence spectra for seven regions equally spaced across entire wafer. Spectra are offset and increase in intensity with growth temperature as indicated.

Figure 3 – PL peak emission energy and PL intensity as a function of position/temperature on the wafer.

Figure 4 – Room temperature photo-modulated transmission spectra for seven regions across the entire region. Arrows indicate bandgap energies used to calculate indium concentrations using Equation (1) for corresponding region.

Figure 5 – Calculated indium concentration in each region using observed bandgaps from PT experiments. Indium concentration, x, was calculated using Equation (1).

Figure 6 – AFM images (1μm x 1μm) of regions 1,4, and 7. (a) Region 1 with a surface roughness RMS of 1.8 nm, (b) Region 4 with a surface roughness RMS of 5.2 nm and (c) Region 7 with a surface roughness RMS of 6.6 nm, are shown. It is important to note that the AFM analysis was performed on the final capping barrier which is ~200 Å of GaN and not on the InGaN layer which only has a thickness of ~30 Å.

Figure 1 of 6

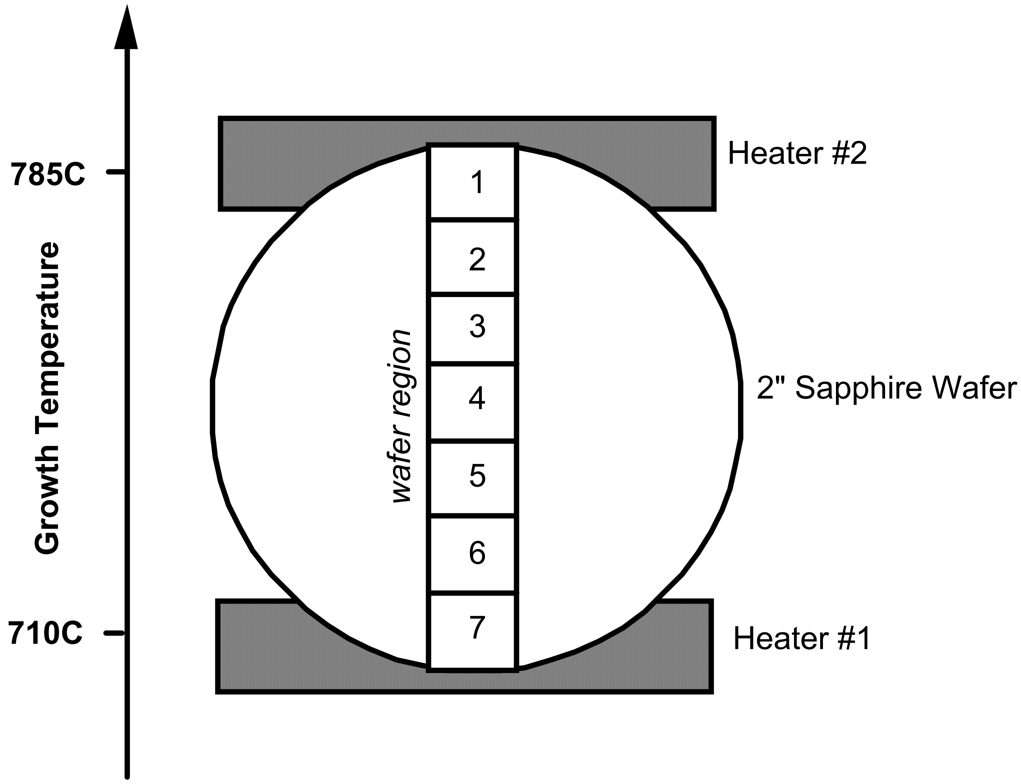

Growth Temperature
785C
710C
Heater #2
1
2
3
4
5
6
7
wafer region
2" Sapphire Wafer
Heater #1

Figure 2 of 6

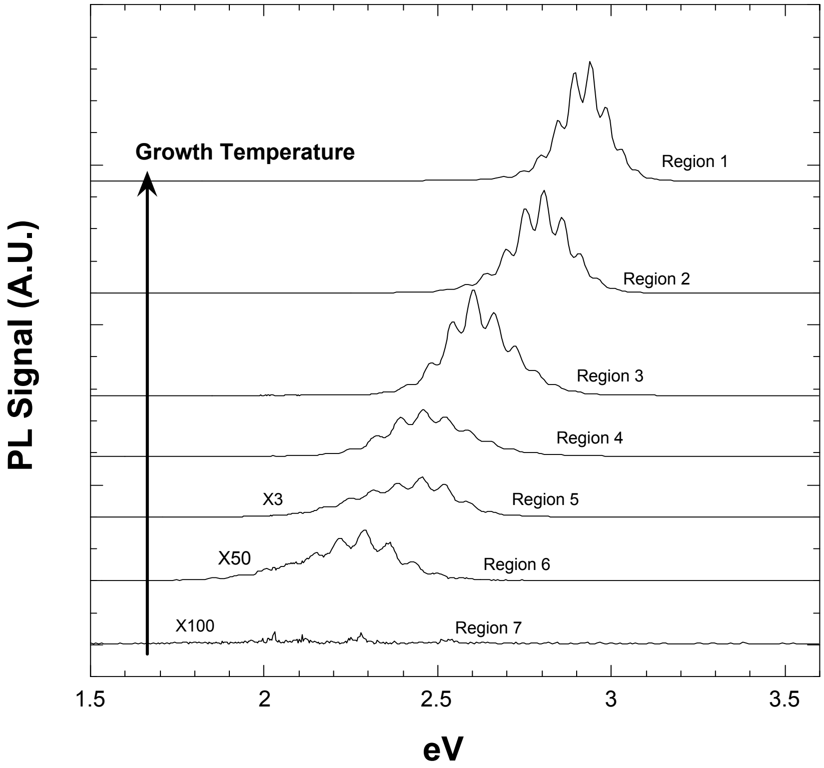

Growth Temperature
Region 1
Region 2
Region 3
Region 4
X3
Region 5
X50
Region 6
X100
Region 7
PL Signal (A.U.)
1.5
2
2.5
3
3.5
eV

Figure 3 of 6

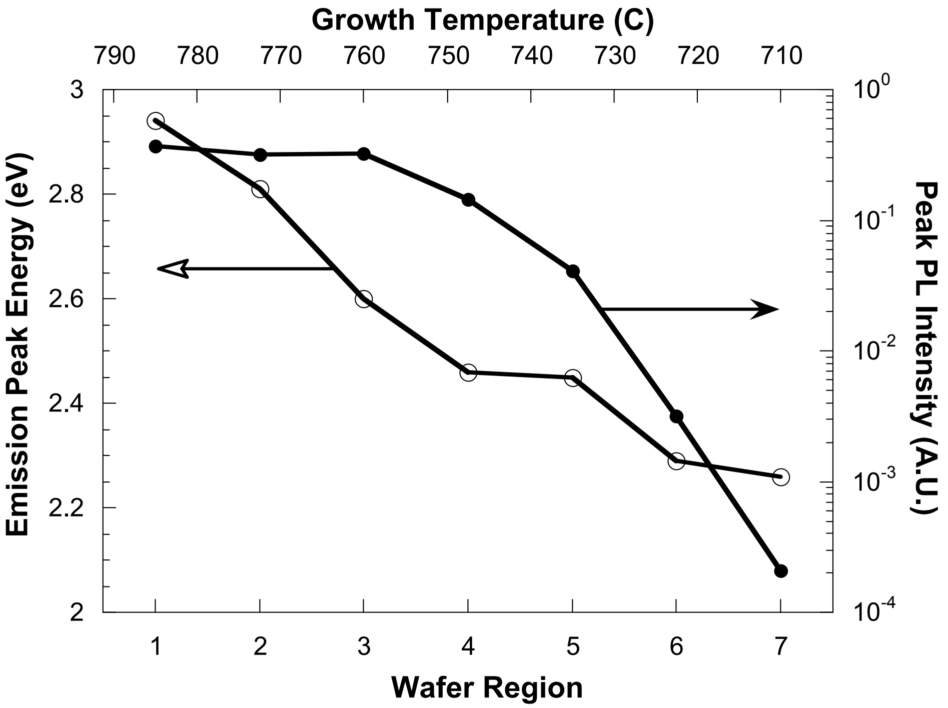

Growth Temperature (C)
790
780
770
760
750
740
730
720
710
3
2.8
2.6
2.4
2.2
2
Emission Peak Energy (eV)
10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
Peak PL Intensity (A.U.)
1
2
3
4
5
6
7
Wafer Region

Figure 4 of 6

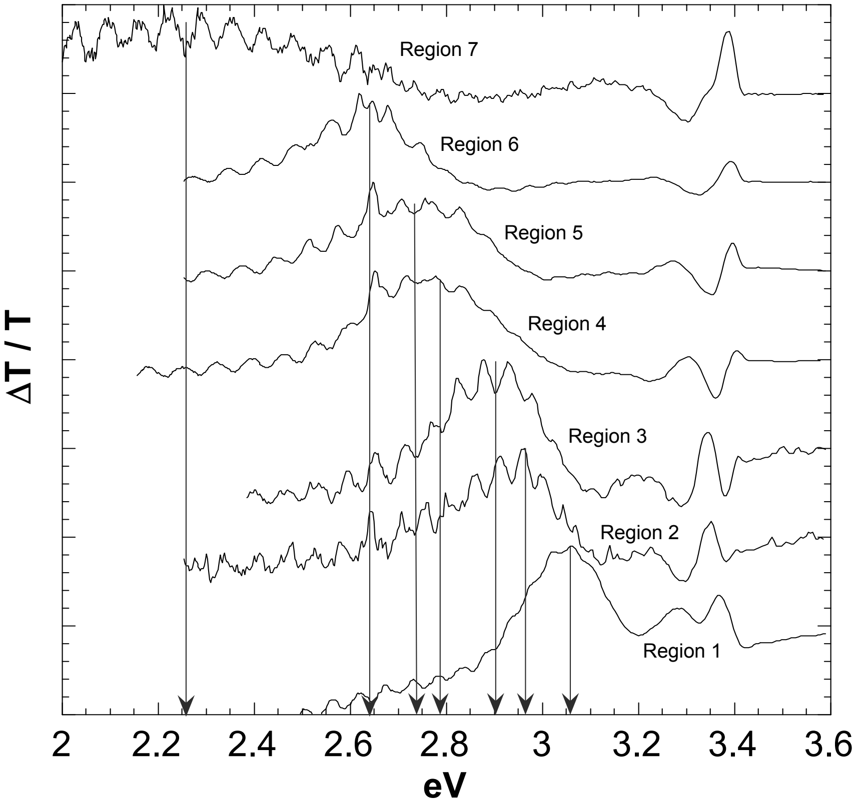
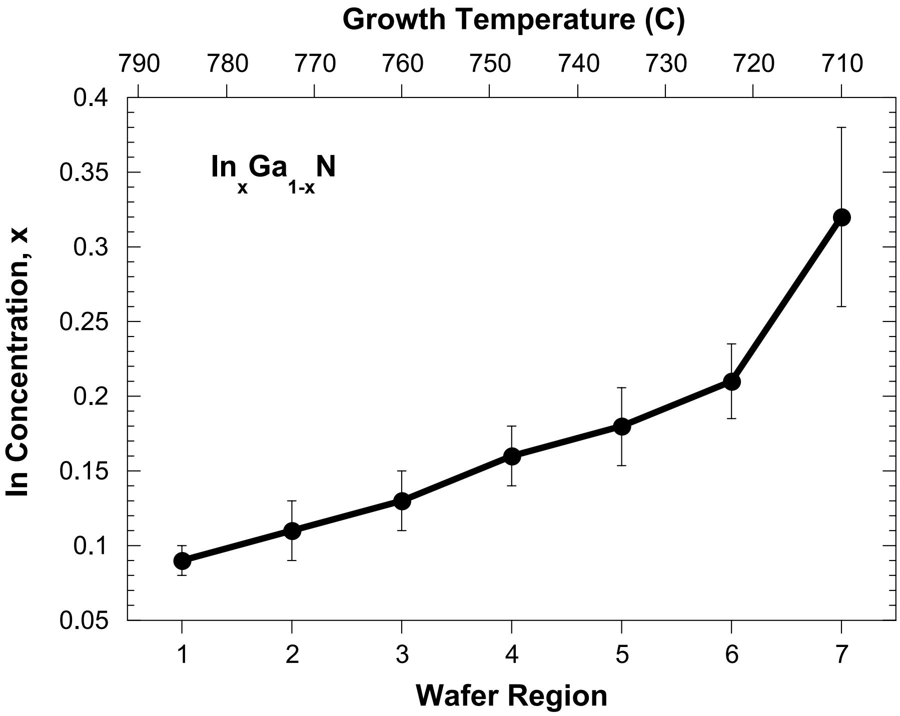

Region 7
Region 6
Region 5
Region 4
Region 3
Region 2
Region 1
ΔT / T
2
2.2
2.4
2.6
2.8
3
3.2
3.4
3.6
eV

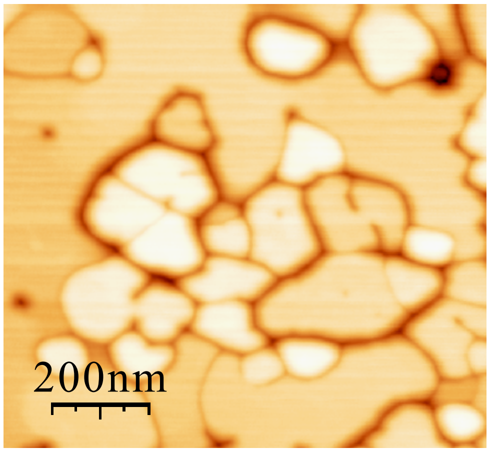
Figure 5 of 6



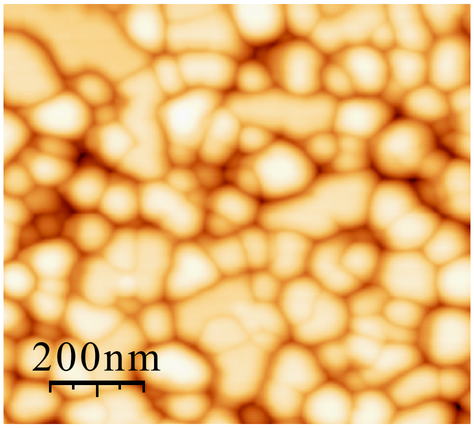
Growth Temperature (C)
790
780
770
760
750
740
730
720
710
0.4
0.35
0.3
0.25
0.2
0.15
0.1
0.05
In Concentration, x
$In_xGa_{1-x}N$
1
2
3
4
5
6
7
Wafer Region

Figure 6 of 6

a) Region 1



b) Region 4



c) Region 7