

Optimization of InGaN/GaN multiple quantum well layers by a two-step varied-barrier-growth temperature method

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Abstract

We report the effect of different temperature profiles on the quality of the InGaN/GaN multiple quantum well (MQW) structures by employing photoluminescence (PL) and atomic force microscopy (AFM). We adopted a two-step varied-barrier-growth temperature method to improve the structural and optical properties of the InGaN/GaN MQW layers. The low-temperature GaN barrier layer was introduced to reduce the desorption rate of the indium atoms of the InGaN well, and then the high-temperature GaN barrier was grown to reduce the defects of InGaN/GaN MQWs. When the width of the low-temperature GaN barrier was 50 Å and the high-temperature GaN barrier was grown at 1000 °C, the defect and surface roughness were significantly reduced, especially with a reduction in the depth of V-defect as low as 20 Å.

1. Introduction

InGaN and InGaN/GaN multiple quantum wells (MQWs) have been used as the active layers for optoelectronic devices such as nitride-based light-emitting diodes (LEDs) and laser diodes (LDs) [1, 2]. The deposition of high-quality InGaN/GaN MQW structures using metalorganic chemical vapor deposition (MOCVD) has become very important in device performance. Despite the surprising advances in device performance, the progress of these devices is often limited by the fundamental problems of InGaN. There is a trade-off between the epilayer quality and the amount of InN incorporation into InGaN as the growth temperature is changed. Lowering the growth temperature resulted in an increase in the allowable InN concentration range, but at the expense of reduced crystalline quality [3]. Therefore, the optimal-growth temperature of InGaN well layers is normally much lower than that of GaN barrier layers. It is difficult to achieve a high-quality InGaN/GaN MQW structure with a constant MQW growth temperature. It is also difficult to

achieve a uniform distribution of indium atoms in the InGaN well layer due to indium desorption or indium segregation of InGaN at the typical growth temperature of GaN. To avoid such a problem, it would be beneficial to grow the InGaN well layers and GaN barrier layers at different temperatures. Furthermore, it has been reported that growth parameters such as growth temperature [4–10], growth pressure [11], growth interruption [12, 13] and the use of H₂ carrier gas [14] significantly affect the crystal quality of InGaN/GaN MQWs and the performance of the nitride-based LEDs. Thus, in order to understand more clearly the effect of growth conditions (temperature and carrier gas, etc) on the crystal quality of InGaN/GaN MQWs, the structural and optical properties of InGaN/GaN MQWs with various growth conditions must be studied in detail.

In addition, a large lattice mismatch and a thermal expansion coefficient difference between GaN and sapphire cause several defects in a GaN layer, such as threading dislocations (TDs), stacking faults and inversion domain boundaries (IDBs) [15]. Especially, it has been reported that TDs disrupt the InGaN/GaN MQWs and initiate the V-defects

[16–18]. The V-defects have been frequently observed in InGaN/GaN MQW structures. These crystal imperfections affect the electrical characteristics of the devices, such as leakage current. The leakage current in InGaN/GaN MQW LEDs is critical for device reliability, lifetime and degradation in high-power operation [19]. Therefore, it is of great importance to optimize the layer structures of InGaN/GaN MQWs for further improved device performance.

In this work, we examine the effect of the growth temperature for GaN barrier layers on the optical and structural properties of the InGaN/GaN MQW layers by using photoluminescence (PL) and atomic force microscopy (AFM). The barrier temperature has a strong effect on the optical properties of the wells through indium desorption and indium segregation. Therefore, we adopted the two-step varied-barrier-growth temperature method, in which the low-temperature GaN barrier layer was introduced to reduce the desorption rate of the indium atoms in InGaN wells, and then the high-temperature GaN barrier layer was grown to reduce the defects of the InGaN/GaN MQW layers.

2. Experimental details

InGaN/GaN MQW structures were grown on c-plane sapphire substrates by MOCVD. Trimethylgallium (TMGa), trimethylindium (TMIn) and ammonia (NH_3) were used as the source precursors for Ga, In and N, respectively. Biscyclopentadienyl magnesium (CP2Mg) and silane (SiH_4) were used as the p-type and n-type doping sources, respectively. The reactor pressure was maintained at 300 Torr throughout the entire process. The temperature of the MOCVD equipment was precisely controlled by using a pyrometer. The structures were composed of a Si-doped, $3\ \mu\text{m}$ thick n-type GaN layer with a carrier concentration of $5 \times 10^{18}\ \text{cm}^{-3}$, five-period MQW active layers consisting of GaN barriers and $20\ \text{\AA}$ thick $\text{In}_{0.2}\text{Ga}_{0.8}\text{N}$ wells. The GaN barrier was grown at two different temperatures. At low temperatures, its thickness was varied from 0 to $70\ \text{\AA}$ to investigate the effect of its thickness on the PL characteristics of MQW layers, while it was fixed to be $75\ \text{\AA}$ at high temperatures. The details of growth conditions are as follows. Prior to the growth, sapphire substrates were first heated to $1100\ \text{^\circ C}$ in hydrogen ambient for 10 min to remove surface contamination, and then cooled down to $560\ \text{^\circ C}$ to grow the GaN-nucleation layer. Next, the temperature was elevated to $1000\ \text{^\circ C}$ to grow a $3\ \mu\text{m}$ thick n-type GaN layer. The substrate temperature was subsequently ramped down to $770\ \text{^\circ C}$ to grow the InGaN QW in the active region. During the growth of the QW layers, N_2 was used as an ambient to increase indium incorporation into the InGaN QW layers. During the growth of the InGaN/GaN MQW layer, different growth conditions were used as follows. Figure 1 shows the schematic of the growth conditions for the MQW layer. Before the growth of the high-temperature GaN barrier layer, we adopted the low-temperature GaN barrier to reduce the desorption rate of the indium atoms. The low-temperature GaN barrier was grown at the same temperature as the InGaN well layer. Then, the high-temperature GaN barrier was grown at the temperature range of $770\text{--}1040\ \text{^\circ C}$ in an ambient of H_2 .

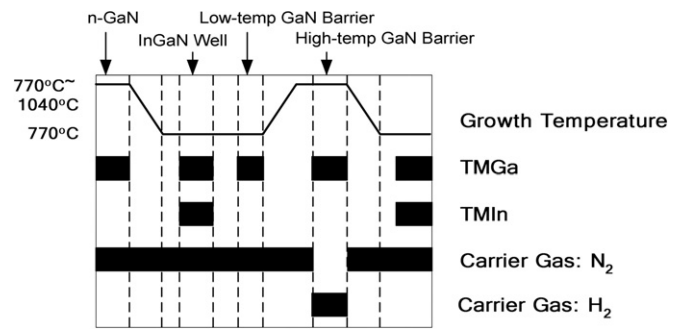


Figure 1. A schematic diagram of the growth procedure used for growing the MQW active regions.

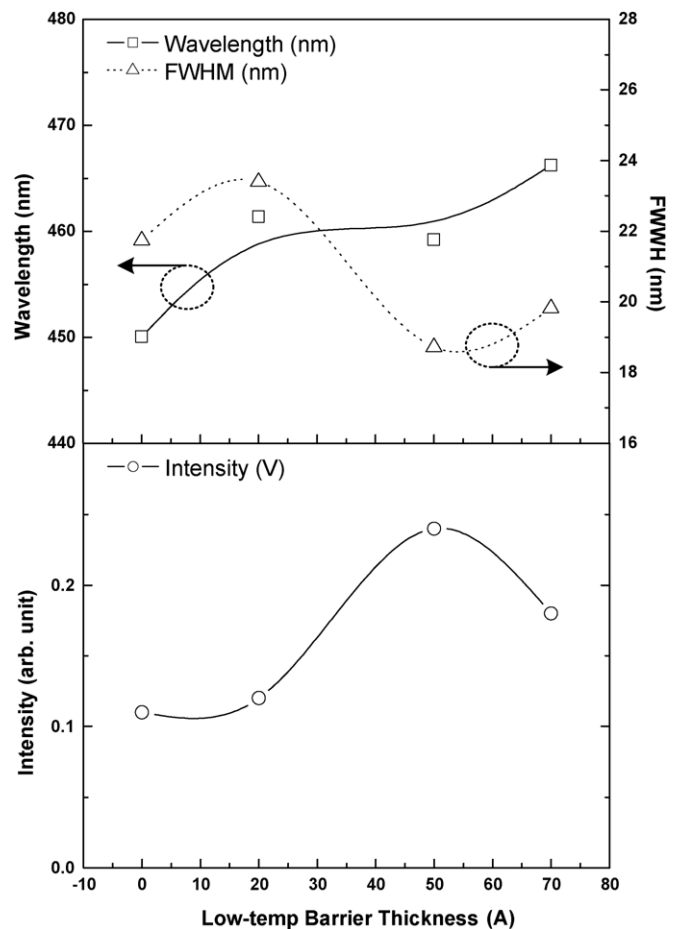


Figure 2. Room-temperature PL peak wavelength, full-width at half-maximum (FWHM) and its intensity as a function of the thickness of the low-temperature GaN barrier layer.

The crystal qualities of these epitaxial layers were evaluated by AFM and PL. PL measurements were conducted at room temperature using the excitation beam of a 325 nm He–Cd laser.

3. Results and discussion

In order to understand the influence of the low-temperature GaN barrier layer on the optical properties of InGaN/GaN MQW heterostructures, five-period InGaN/GaN MQW

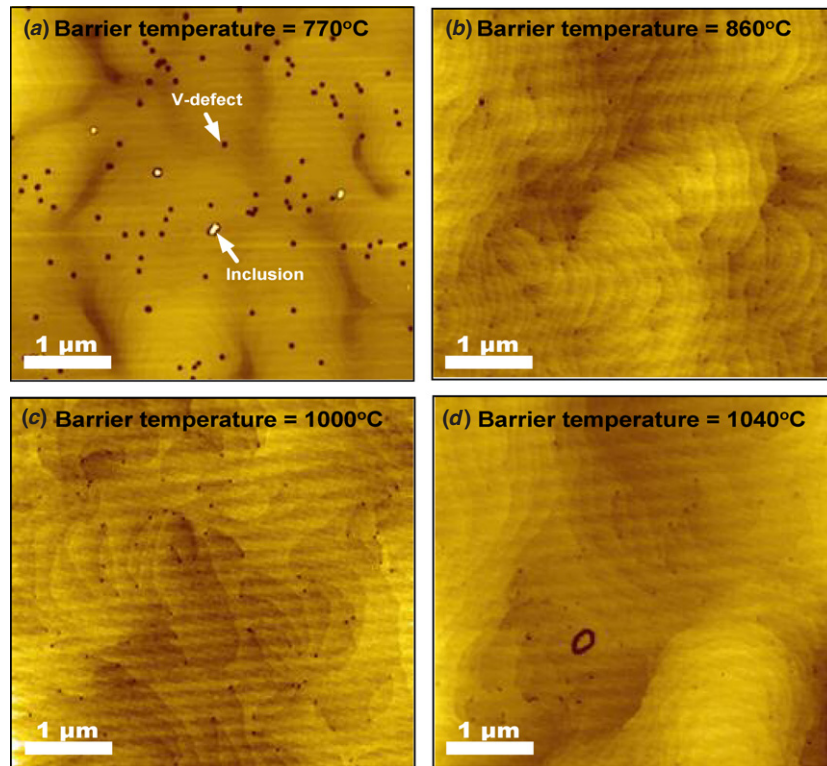


Figure 3. AFM surface morphology scans (the $5\ \mu\text{m} \times 5\ \mu\text{m}$ scale) of the MQW layers grown at four different barrier-growth temperatures: (a) $770\ ^\circ\text{C}$, (b) $860\ ^\circ\text{C}$, (c) $1000\ ^\circ\text{C}$ and (d) $1040\ ^\circ\text{C}$.

(This figure is in colour only in the electronic version)

structures were grown with different thicknesses of the low-temperature GaN barrier layers. The growth temperature of the high-temperature GaN barrier layer was kept constant at $1000\ ^\circ\text{C}$ after the growth of the low-temperature GaN barrier layer. Figure 2 shows the room-temperature PL peak wavelength, full-width at half-maximum (FWHM) and its intensity measured from the InGaN/GaN MQWs as a function of the thickness of the low-temperature GaN barrier layer. When the barrier thickness was $0\ \text{\AA}$ (that is, without the low-temperature GaN barrier), the peak emission wavelength was observed at $450\ \text{nm}$. However, it was red-shifted to $461.3\ \text{nm}$ and $459.2\ \text{nm}$, respectively, for InGaN/GaN MQWs with $20\ \text{\AA}$ and $50\ \text{\AA}$ thick GaN barrier layers. As the thickness of the barrier was increased to $70\ \text{\AA}$, PL peak wavelength showed a large red-shift up to $467\ \text{nm}$. We suggest that the InGaN well layer might be desorbed during the growth interruption for temperature ramping. As mentioned above, as the thickness of the low-temperature GaN barrier was decreased, PL peak wavelength was blue-shifted. Thus, the blue shift of PL wavelength indicates that the well layer was desorbed as the thickness of the low-temperature GaN barrier was reduced. Therefore, we interpret that the InGaN well layer is desorbed while the temperature is ramped up. In addition to the wavelength shift caused by the desorbed InGaN well layer, it could be also thought that indium content in the InGaN well might be decreased as the thickness of the low-temperature GaN barrier is decreased. PL energy in InGaN/GaN QW is affected by the quantum confined Stark effect (QCSE), which is mainly induced by piezoelectric polarization and is sensitive

to the strain of the InGaN layer. Therefore, the electric field across QW may be weakened by decreasing the strain of QW and the effective transition energy in QW would be increased due to the weakening of QCSE [20]. We also investigated the FWHM and the intensity of PL to know the optical quality of InGaN/GaN MQW layers. When the thickness of the low-temperature barrier was $50\ \text{\AA}$, the intensity became strong and the FWHM decreased to $18\ \text{nm}$. Therefore, we set the thickness of the low-temperature barrier to $50\ \text{\AA}$ in this experiment.

The high-temperature GaN barrier layer was grown at different temperatures to further understand its effect on the structural and optical properties of the MQW layers. While $75\ \text{\AA}$ thick high-temperature GaN barrier layers were grown at various growth temperatures from $770\ ^\circ\text{C}$ to $1040\ ^\circ\text{C}$, the thickness of the low-temperature GaN barrier layer was kept constant at $50\ \text{\AA}$. Figure 3 displays the AFM surface morphology scans (in the $5\ \mu\text{m} \times 5\ \mu\text{m}$ scale) of the MQW layers grown at four different barrier-growth temperatures. For a barrier temperature of $770\ ^\circ\text{C}$, which is the same temperature as the low-temperature GaN barrier, large size of pits was observed while the other samples showed small size of pits. The shape of pits is an inverted hexagonal pyramid called a ‘V-defect’ in InGaN/GaN MQWs [7]. Furthermore, inclusions were also seen in the sample for the barrier temperature of $770\ ^\circ\text{C}$ (figure 3(a)). However, inclusions were not seen in the samples of which GaN barriers were grown at elevated temperature, as shown in figures 3(b)–(d). Inclusions were known to play a detrimental role, limiting the

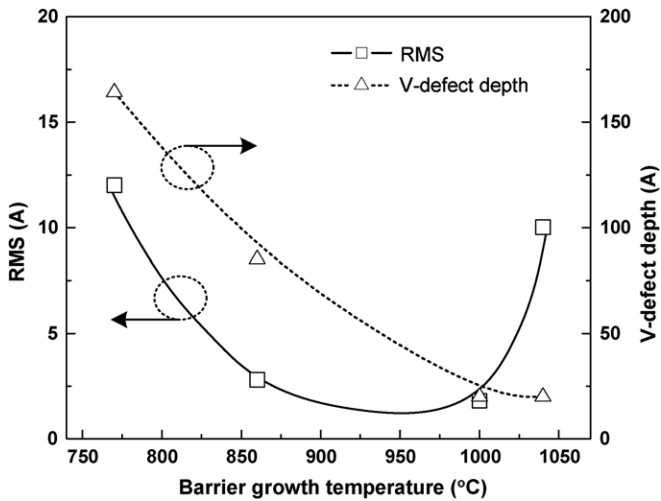


Figure 4. Root-mean-square (RMS) values and the depth of V-defects as a function of the growth temperature for the high-temperature GaN barrier layer.

thermal stability of the LED devices [21, 22]. Therefore, when the GaN barrier-growth temperature is the same as that of the InGaN well (that is, 770 °C), InGaN/GaN MQW layers were deteriorated due to the enlargement of V-defect and inclusions.

Figure 4 shows the measured root-mean-square (RMS) values and the depth of V-defects of MQWs grown at various barrier-growth temperatures. RMS values were obtained in $1 \mu\text{m} \times 1 \mu\text{m}$ scan areas. When the GaN barrier-growth temperature was the same (770 °C) as that of the InGaN well, the RMS value and depth of V-defects were as large as 12 Å and 164 Å, respectively. On the other hand, as the temperature increased, the surface roughness and depth of V-defects decreased. The result clearly showed that at the barrier temperature of 1000 °C, the RMS value and depth of V-defects significantly decreased to 1.8 Å and 20 Å, respectively. However, the surface roughness (RMS value) was increased to 10 Å as the barrier-growth temperature was further increased to 1040 °C. Consequently, the defects and surface roughness were significantly reduced when the width of the low-temperature GaN barrier layer was 50 Å and the high-temperature GaN barrier layer was grown at 1000 °C, especially with a reduction of the depth of V-defects as low as 20 Å.

We also performed PL measurements at room temperature to evaluate optical properties of the MQWs with the high-temperature GaN barrier grown at various growth temperatures. As shown in figure 5, the PL peak wavelength for samples of the barrier-growth temperature of 770 °C, 860 °C and 1000 °C was 470, 472.2 and 476.6 nm, respectively. The PL peak wavelength and FWHM were slightly increased until 1000 °C. However, the wavelength was dramatically decreased to 446.4 nm for the barrier temperature of 1040 °C. In addition, PL intensity was not decreased until 1000 °C while PL intensity for the barrier temperature of 1040 °C was decreased. In general, the high growth temperature of the GaN barrier limits the maximum indium content and wavelength accordingly, while the high growth temperature is needed for

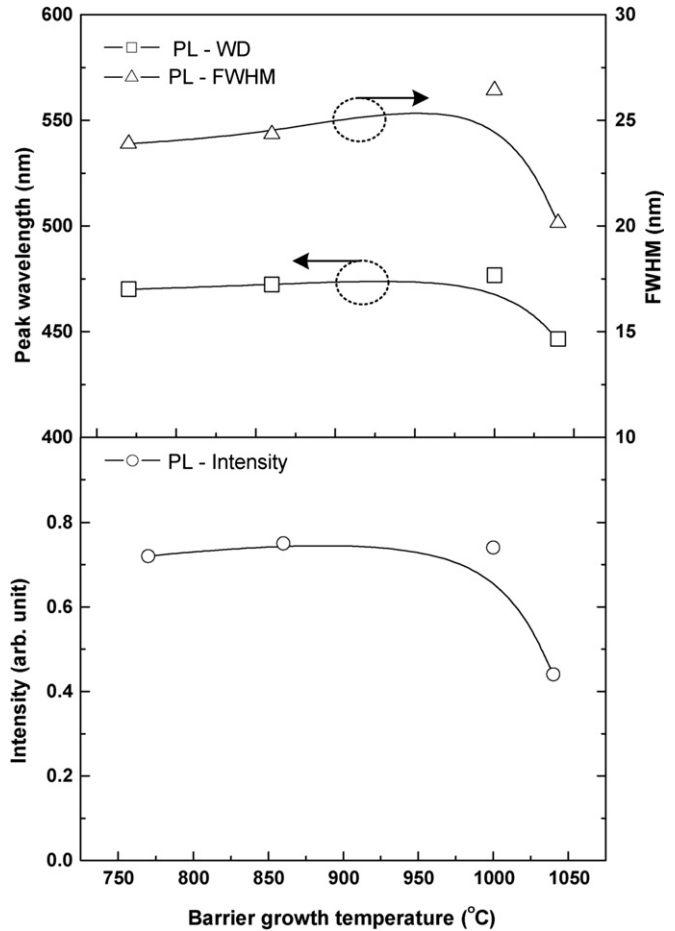


Figure 5. Room-temperature PL peak wavelength, full-width at half-maximum (FWHM) and intensity as a function of the growth temperature for the high-temperature GaN barrier layer.

high quality of the InGaN/GaN MQWs. In this work, we determined the optimized barrier-growth temperature from the results of AFM and PL.

4. Conclusions

We investigated the effect of two-step varied-barrier-growth temperature profiles on the quality of the InGaN/GaN MQW structures. Their optical and structural properties were investigated by employing PL and an AFM. The barrier temperature had a strong effect on the optical properties of the wells through indium desorption and indium segregation. The thickness of the low-temperature GaN barrier was investigated to reduce the desorption rate of the indium atoms of the InGaN well, and then the growth temperature of the high-temperature GaN barrier was examined to reduce the defects of InGaN/GaN MQWs. When the width of the low-temperature GaN barrier was 50 Å, the peak wavelength was 469.2 nm, the intensity became strong and the FWHM decreased to 18 nm. When the high-temperature GaN barrier was grown at 1000 °C, the defects and surface roughness were significantly reduced. Especially the depth of V-defect was reduced to as low as 20 Å.

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