Optical Properties of A-Plane InGaN/GaN Multiple Quantum Wells Grown on Nanorod Lateral Overgrowth Templates

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***Abstract*— A-plane InGaN/GaN multiple-quantum wells (MQWs) were grown on a series of nanorod epitaxial lateral overgrowth (NRELOG) templates with varied nanorod depth. Optical properties of these samples were investigated by excitation power and temperature-dependent photoluminescence (PL). Due to the absence of quantum-confined Stark effect, the negligible PL emission peak shift and nearly identical power index for all samples were observed. In contrast to the as-grown MQWs, the thermal activation energy and internal quantum efficiency of NRELOG MQWs exhibit 1.6-fold and 4-fold increases, respectively, which are attributed to the improvement of crystal quality by NRELOG. Furthermore, the Shockley-Read-Hall nonradiative coefficient, determined from the fits of power-dependent PL quantum efficiency, is also apparently reduced while MQWs are grown on NRELOG GaN template. The results show the feasibility to fabricate high radiative efficiency a-plane devices via NRELOG.**

***Index Terms*— A-plane, InGaN/GaN multiple quantum wells, internal quantum efficiency, nanorod lateral epitaxial overgrowth.**

I. INTRODUCTION

THE III-nitride material has attracted much attention due to its tremendous potential for fabricating LEDs with an emission range from UV to visible wavelength [1], [2]. Recently, III-nitride LEDs have been widely utilized in various applications, including intelligent interior lighting, backlighting units for liquid crystal display, and general lighting [3]. In general, the internal quantum efficiency was used to evaluate LED performance and was defined as the generated photon number divided by the injected carrier numbers. From the viewpoint of energy-saving, the LED structure must be designed to yield high internal quantum efficiency. However, the conventional c-plane nitride-based quantum wells suffer the quantum-confined Stark effect as a result of the existence of spontaneous and piezoelectric polarization fields that are parallel to [0001] c direction [4], [5].

This effect results in spatial separation of the electron and hole wave functions in the quantum wells, which lowers the carrier recombination probability, reduces internal quantum efficiency, and induces red-shifted emission [6]. To avoid such polarization effects, growth along $[11\bar{2}0]$-oriented direction has been explored for planar a-plane GaN prepared on r-plane sapphire [7] by metalorganic chemical vapor deposition (MOCVD). Since these GaN surfaces contain an equal number of Ga and N atoms in each monolayer, electric field free and nonpolar characteristics are obtained. The recent studies of a-plane InGaN/GaN multi-quantum wells (MQWs) demonstrate that it is possible to eliminate such polarization fields along the nonpolar orientation [8], [9]. However, the difficult issue to utilize nonpolar GaN is that no suitable substrate can be used for hetero-epitaxial a-plane GaN growth. In general, a threading dislocation (TD) density of $\sim 3\times10^{10}\text{cm}^{-2}$ and on-axis x-ray diffraction full width at half maximum (FWHM) of ~0.3° were commonly observed in a-plane GaN grown on r-plane sapphire because of the large anisotropic lattice mismatch between these two materials [7]. The TDs in GaN act as nonradiative recombination centers and charge scattering centers which are responsible for poor internal quantum efficiency and low carrier mobility [10], [11]. Therefore, the reduction of TD density is essential to improve the performance of a-plane light-emitting devices. Lateral epitaxial overgrowth (LEO) techniques were widely employed in the past to reduce defect density in nonpolar GaN [12]. In our previous work, we performed the LEO on a series of nanorod templates with varied etching depth to realize the defect-reduction and quality improvement in the subsequently grown a-plane GaN layer [13]. The average TD density can be reduced from 3×10^{10} to $1\times10^{9}\text{cm}^{-2}$. The x-ray diffraction FWHMs for on-and off-axis reflections were also decreased from 1480 to 514 arc sec and from 2420 to 1232 arc sec, respectively. This demonstrated that one can achieve better crystal quality a-plane GaN using nanorod epitaxial lateral overgrowth (NRELOG).

Up to now, the structural and optical properties of a-plane InGaN/GaN MQWs are still much inferior compared to the mature c-plane counterpart due to very high TD density while

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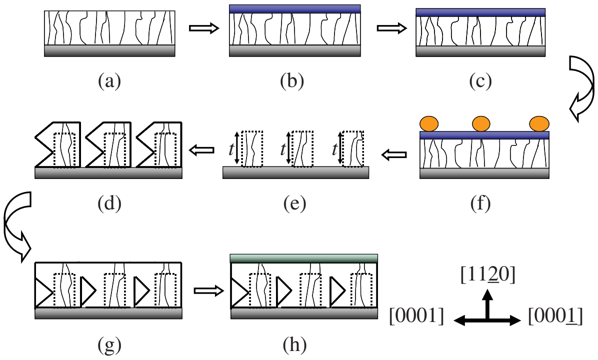


Fig. 1. Schematic flowchart of a-plane GaN NRELOG process. (a) a-GaN template. (b) 200 nm SiO_2 deposition. (c) 10 nm Ni deposition. (d) Formation of Ni particles by thermal annealing. (e) Nanorods formed by RIE/ICP (vary etching depth "t" from 0.2 μm to 1.7 μm). (f) MOCVD regrowth. (g) Fully coalesced NRELOG template. (h) MQWs deposition.

grown on r-plane sapphire substrates. Therefore, in the paper, we deposit InGaN/GaN multiple quantum wells (MQWs) on the high crystal quality NRELOG a-plane GaN templates and investigated their optical properties as well as internal quantum efficiency by excitation power and temperature dependent photoluminescence (PL).

II. EXPERIMENTS

The schematic flowchart of NRELOG is shown in Fig. 1. First, a 1.7-μm-thick a-plane GaN layer was grown on r-plane sapphire by MOCVD, followed by the deposition of a SiO_2 film with a 200 nm thickness and a Ni film with a 10 nm thickness acting as etching masks. Subsequently, a rapid thermal annealing treatment of 850 °C was utilized to obtain nano-scale Ni masks. Then, the GaN nanorods were etched through the nano-mask openings by reactive ion etching (RIE)/inductively coupled plasma etching (ICP). After that, the residual SiO_2 masks on nanorods were removed by hydrofluoric acid to simplify the growth process. From SEM observation (now shown here), the diameter of the nanorod is 300 $\sim$ 500 nm and the nanorod density is estimated to be around $6\times10^8/\text{cm}^2$. In order to optimize the quality improvement in the regrown a-plane GaN layer, we designed a series of nano-rod templates with different etching depths (t) from 0.2 μm to 0.7 μm, 1.2 μm, and 1.7 μm. Then, the GaN regrowth was performed by MOCVD on these nanorod templates. After the fully coalesced NRELOG GaN templates are achieved, we subsequently deposit InGaN/GaN MQWs on these templates, which comprise 6 pairs of 7 nm $In_{0.2}Ga_{0.8}N$ well and 20 nm GaN barrier, and then the excitation power and temperature dependent PL measurement is carried out to investigate optical properties as well as internal quantum efficiency. For excitation power and temperature-dependent PL measurement, these MQWs sample were mounted in a closed-cycle He cryostat and the temperature was controlled over the range from 20 to 300 K. A 325 nm cw He-Cd laser was used as the pumping source and the exciation power density was changed from 0.1 to 280 W/cm^2. The emitted luminescence light was collected through a 0.32 m spectrometer with a charge-coupled device detector.

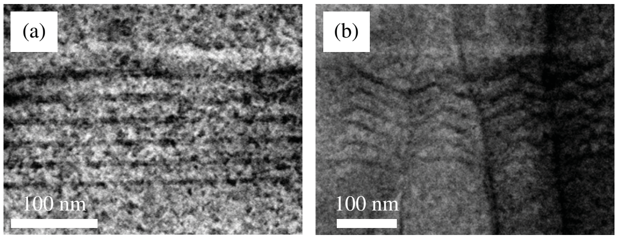


Fig. 2. Cross-sectional TEM image of a-plane MQWs grown on (a) NRELOG GaN template and (b) as-grown GaN template.

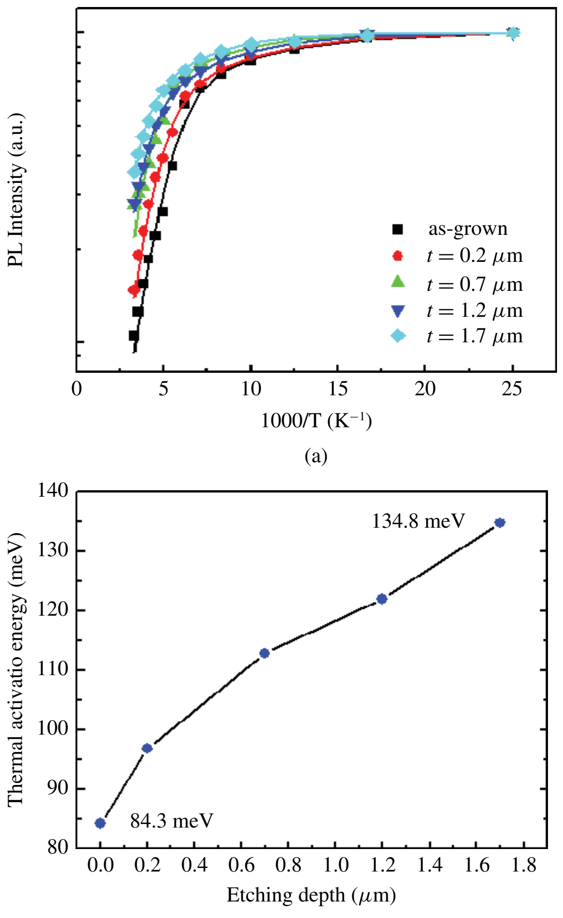
III. RESULTS AND DISCUSSION

In our previous work [13], by utilizing AFM scanning, x-ray diffraction measurement, and TEM observation, we have demonstrated that the crystal quality of a-plane GaN NRELOG films can be gradually improved with the increase in the etching depth (t) of the nanorod templates. In order to observe the microstructure of the a-plane MQWs grown on NRELOG GaN template and as-grown GaN template, the typical bright-field cross-sectional TEM image near $[1\underline{1}00]_{\text{GaN}}$ zone axis is performed as shown in Fig. 2. Fig. 2(a) presents the morphology of MQWs grown on NRELOG GaN template and Fig. 2(b) shows the morphology of MQWs grown on as-grown GaN template. In contrast to Fig. 2(b), the NRELOG MQWs exhibits flat and clear boundaries between InGaN wells and GaN barriers as a result of the elimination of threading dislocations in lateral overgrowth region. It should be noted that the morphology of as-grown MQWs is much inferior compared to NRELOG MQWs, the boundaries between InGaN wells and GaN barriers were severely interfered by many TDs that would give rise to the poor carrier confinement and low internal quantum efficiency.

It has been studied that the carrier could receive activation energy to thermalize from the radiative or localized centers, to nonradiative or delocalized centers as the temperature is increased [14]. In other words, the temperature-induced quenching of luminescence is related to the thermal activation of photogeneratd carriers from nonradiative defect states, or the thermal excitation of carriers out from confined quantum-well states to barrier states. Therefore, it is expected that the MQWs with better confinement should exhibit slower thermal quenching of luminescence and have larger activation energy. In order to further verify that, the experimental temperature-dependent PL data were fitted by following Arrhenius equation to investigate the carrier behavior during the thermal processes [15]

$$I\ (T) = \frac{I_0}{1 + A * \exp\left(-\frac{E_a}{K_B T}\right) + B * \exp\left(-\frac{E_b}{K_B T}\right)}$$

where $I(T)$ is the temperature-dependent PL intensity, I_0 is the PL intensity at 20 K, K_B is Boltzmann's constant, A and B are the rate constants, and E_a and E_b are the activation energies for two different nonradiative channels, which can be distinguished for the low temperature and high temperature regions [16]. It is obvious that as-grown MQWs exhibits fastest roll-off of PL intensity with increasing temperature,



(a)

(b)

Fig. 3. (a) Normalized PL intensity plotted as a function of $1/T$ for the as-grown MQWs and NRELOG MQWs with different nanorod etchig depth. The symbols stand for the measurement results and the solid lines mean the fitted curve of these five samples. (b) Fitted activation energy as a function of nanorod etchig depth, where 0 μm etching depth represents the as-grown sample.

as shown in Fig. 3(a), which can be attributed to that the as-grown MQWs are severely interfered by lots of TDs. The fitted activation energy of these MQW samples is listed in Fig. 3(b). Since the crystal quality of a-plane GaN NRELOG films are gradually improved with nanorod etching depth, the thermal activation energy gradually increases and reaches the maximum value of 134.8 meV when grown on 1.7 μm etching depth of NRELOG GaN template. It is believed the activation energy obtained from the PL intensity reflects the effective energy barrier to nonradiative recombination centers, which relate to the crystal quality and defect density. Thus, the activation energy will be enhanced with increasing etching depth, due to the improvement of the GaN template crystal quality.

The internal quantum efficiency is one of the most important factors to limit the MQW performance. The conventional approach to characterize the internal quantum efficiency is to compare the PL intensity between the low and room temperatures; however, the selected power density of excitation would profoundly affect the measured results. Therefore, in order to evaluate internal quantum efficiency more accurately, we performed the excitation power density and temperature-dependent PL measurement to determine the internal quantum efficiency [17], [18]. The power-dependent measurement results for the as-grown and the MQWs grown on the different depth NRELOG GaN templates have been summarized and plotted as a function of the excitation power density in Fig. 4. The efficiency is defined as the collected photon numbers divided by the injected photon numbers and all normalized to the maximum efficiency. For all samples in this investigation, the internal quantum efficiency increases distinctly while the excitation power density further increases and reaches its maximum value. It is associated with the fact that the radiative recombination rate is gradually dominated while the injected power density increases. These efficiency curves at 15 and 300 K reveal similar inclination respectively. The corresponding density of the peak efficiency at 300 K is at injected power density of about 100 W/cm^2. In terms of the peak efficiency at 300 K, the internal quantum efficiency (∼40%) of MQWs grown on 1.7 μm etching depth of NRELOG GaN template was significantly enhanced in contrast to that of as-grown MQWs (10%). It means that under the same excitation power density, there is about 4-fold enhancement for the converted photon numbers within the MQW active region while compared to the as-grown MQW sample. Besides, with regard to the phenomenon of the efficiency droop at low-temperature, we suggest that when injected carriers keep increasing, the filling of localized states will make carriers more easily escape from localized states to extended states, resulting in little deterioration of the internal quantum efficiency. Based on our experimental results, we think the higher internal quantum efficiency for the MQWs grown on NRELOG templates is due to the better crystalline quality, which is consistent with the TEM observation of Fig. 2. The optical performance reveals an obvious enhancement with increasing etch depth of NRELOG GaN templates. MQWs grown on the relatively deep NRELOG GaN template exhibit better performance. We believe that GaN nanorod template should have an optimum depth to achieve the best optical performance. Further investigation on the optimum

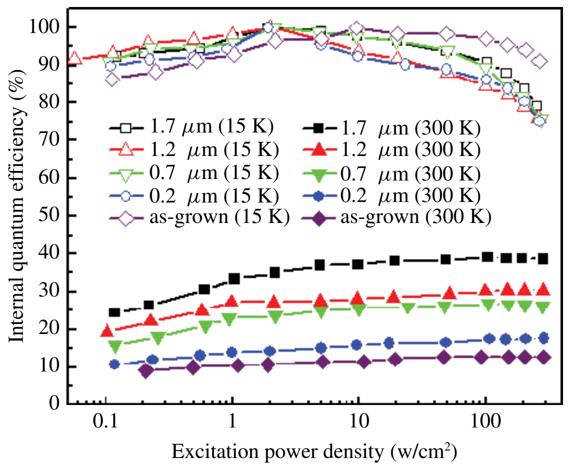
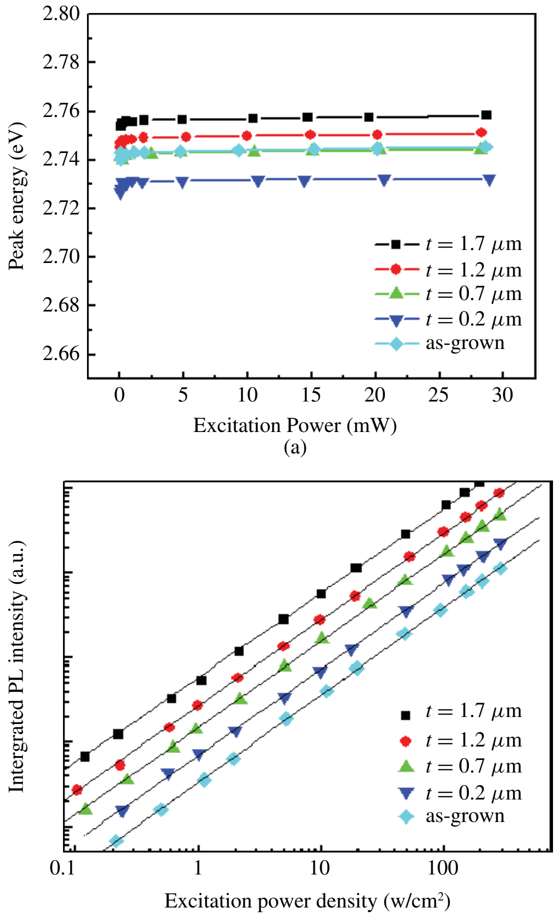


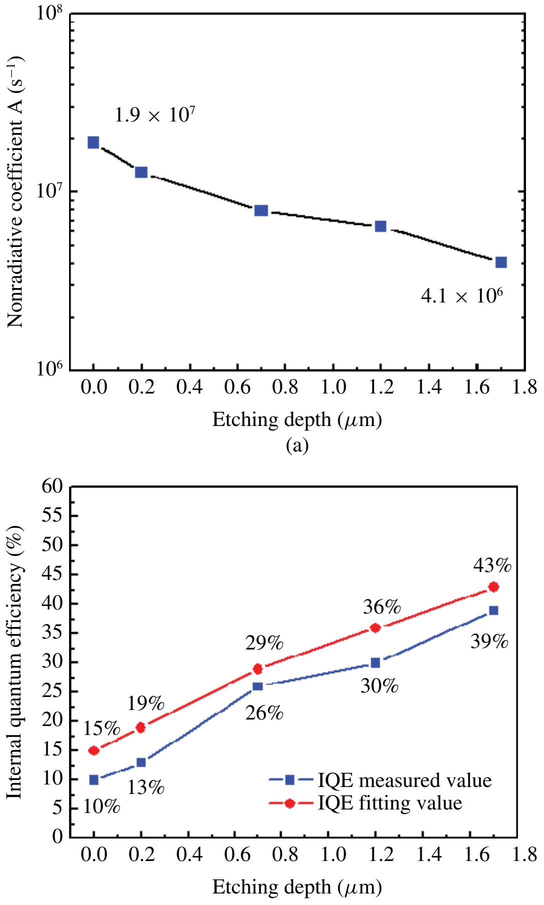
Fig. 4. Internal quantum efficiency as a function of excitation power density at 15 and 300 K for the MQWs grown on as-grown GaN template and the NRELOG GaN templates with different nanorod etching depth.



(a)

(b)

Fig. 5. Power-dependent PL measurement of as-grown MQWs and NRELOG MQWs with different nanorod etchig depth. (a) Emission wavelength versus the pumping power. (b) PL intensity versus the pumping power density and fitted based on the relation $I \sim P^{\alpha}$.



(a)

(b)

Fig. 6. (a) Dependence of nonradiative recombination coefficient on the as-grown MQWs and NRELOG MQWs with different nanorod etchig depth. (b) Simulated and experimental IQE as a function of the different nanorod etching depth for NRELOG MQWs were summarized.

depth shall be carried out in the future. The dependences of PL peak energy-shift on excitation power density are also obtained from the power-dependent PL measurement at room temperature, as shown in Fig. 5(a). All samples show extremely small shift in emission peak energy. The integrated PL intensity was fitted as well, based on the relation $I \sim P^{\alpha}$, where I is the integrated PL intensity, P is the excitation power density, and α is the power index. Fig. 5(b) is the fitting results of integrated PL intensity and the power index α for all samples are around 1. From the negligible PL peak energy shift and nearly identical α index, it suggests that no built-in electric field exists within these a-plane MQW samples [19].

Since crystalline quality of a-plane MQWs is apparently improved using NRELOG, it is expected that the carrier recombination dynamics between as-grown and NRELOG should be quite different. In order to verify that, a method similar to the one proposed in Ref. [20] has been used. We first assume that at steady state the generation rate (G) equals to the total recombination rate (R), including Shockley-Read-Hall nonradiative recombination (An), radiative recombination (Bn^2), and Auger nonradiative recombination (Cn^3), where A, B, C are respective recombination coefficients and n is the carrier density. The integrated PL intensity can be expressed as $I_{\mathrm{PL}} = \eta B n^2$, where η is a collection factor related to the collection efficiency of luminescence and the volume of the excited active region. The coefficient B for nonpolar GaN orientation is generally assumed as 1×10^{-11} cm^3 s^{-1} [21]. In addition, the Auger recombination affects the internal quantum efficiency only at very high excitation. Therefore, the Auger recombination was nearly neglected here. Fig. 6(a) plots the dependence of nonradiative coefficient A on the as-grown MQWs and NRELOG MQWs with different nanorod etching depth. The nonradiative coefficient $A$is apparently reduced to 4.1×10^6 s^{-1}while the MQWs is grown on 1.7 μm etching depth of NRELOG GaN template. Since the threading dislocations have been proved to be nonradiative recombination centers, the reduction of value A is a reasonable consequence of crystal quality improvement by NRELOG. Base on this method, the calculation of the internal quantum efficiency is in good agreement with the experimental results, as shown in Fig. 6(b).

IV. CONCLUSION

In conclusion, we have performed the excitation power density and temperature dependent PL measurement to investigate the optical properties of a-plane MQWs grown on different crystal quality templates. No obvious emission peak shift is observed for all samples, which is evidence of the absence of QCSE. Thermal activation energy for NRELOG MQWs exhibits a higher value (134.8 meV), which can be attributed to the better ability to confine carriers within radiative centers. Under the same injected power density, the internal quantum efficiency in NRELOG MQWs reaches the maximum value of approximately 40%, compared to 10% for as-grown MQWs. The nonradiative coefficient fitted from power-dependent PL data is apparently reduced while grown on NRELOG templates, which is due to elimination of TD density. These results show the potential to fabricate high performance a-plane light-emitting devices via NRELOG.

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