The Effect of RF-Plasma Power on the Growth of III-Nitride Materials

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Abstract: In this study, n-InGaN nanorods were grown directly on p-type Si (111) substrates by plasma-assisted molecular beam epitaxy (PA-MBE). The crystal structure is investigated using the reflection of high-energy electron diffraction patterns. Additionally, the morphology and optical properties of the InGaN nanorods were investigated using both scanning electron microscopy and room temperature photoluminescence spectra. The results showed that, the PL peak position shifted toward lower energy by increasing plasma power due to increase in In-concentration within InGaN nanorods. It can be observed that, through using optimum growth conditions, a uniform indium up to 34% can be achieved with no phase split-up or indium isolation. Thus, none of the mobile In-droplets on the surface were observed.

Keywords*: *n-type InGaN Nanorods, PA-MBE, Photoluminescence Spectra, Plasma Power

1. Introduction

The wurtzite Indium Gallium Nitride InGaN is an efficient substance for photovoltaic and optoelectronic devices due to adjustable band-gap energy which covers from InN (0.70 eV) to GaN (3.4 eV) depending on the amount of indium content (Jani, Ferguson, Honsberg, & Kurtz, 2007). Growth of high quality InGaN single crystal with entire indium composition is extremely challenging due to lattice mismatch and low solubility between InN and GaN atoms causing phase miscibility gap (Doppalapudi, Basu, & Moustakas, 1999). However, thermal expansion coefficient and strain generation at the interface between InGaN and substrate are other difficulties for growing high quality InGaN layer, and also they result in high defects and dislocation density ($\sim 10^{10}$ cm^{-2}) (Goodman *et al.*, 2011; Tabata, Paek, Honda, Yamaguchi, & Amano, 2012). To overcome these difficulties, improving the structure quality and optical properties of the InGaN materials, both nanowires and nanorods, are the optimum choice to overcome all these obstacles. In addition, some studies stated that, growing nanorods offer high crystalline structure associated with almost free strain (Cao *et al.*, 2009; Keating, Urquhart, McLaughlin, & Pearce, 2012).

Recently, a number of researchers reported the growth of InGaN structure with different In concentration by optimizing the growth parameters such as growth temperature and III/V ratio (Dimakis *et al.*, 2005; O'Steen, Fedler, & Hauenstein, 1999). Dimakis *et al.* (2005) found that the

InGaN must be grown at low temperature (530-550 ^{o}C) due to InN decomposition at higher temperature, on the other hand, some of the researchers used the AlN, InN, and GaN buffer layer to improve strain and lattice mismatch between InGaN and the substrate (Cui, Fathololoumi, Kibria, Botton, & Mi, 2012; Shen, Ide, Shimizu, Hara, & Okumura, 2000; Tourbot *et al.*, 2012). Many techniques such as MOCVD, HVPE, and MBE (Johnson *et al.*, 1997; Kim *et al.*, 2004) have been adopted to grow III-nitrides materials. In this research, plasma assisted molecular beam epitaxy (PA-MBE) was used to investigate the effect of RF-plasma power on InGaN growth on Si- substrate.

2. Experimental Procedures

The PA-MBE used in this work in order to provide energetic nitrogen species and standard Knudsen cells such as a Ga and In source. All samples have been grown on the p-type Si (111) substrate without any catalyst. The Si substrates were cleaned first by using solvents such as acetone, methanol, and DI water, then the native oxides were removed by using HF solution. Also, extra cleaning was prepared by in-*situ* thermal annealing process at 800^{o}C in ultrahigh vacuum chamber for 60 min. The reflection high-energy electron diffraction (RHEED) has been used to see a reconstructed Si (7 × 7) patterns.

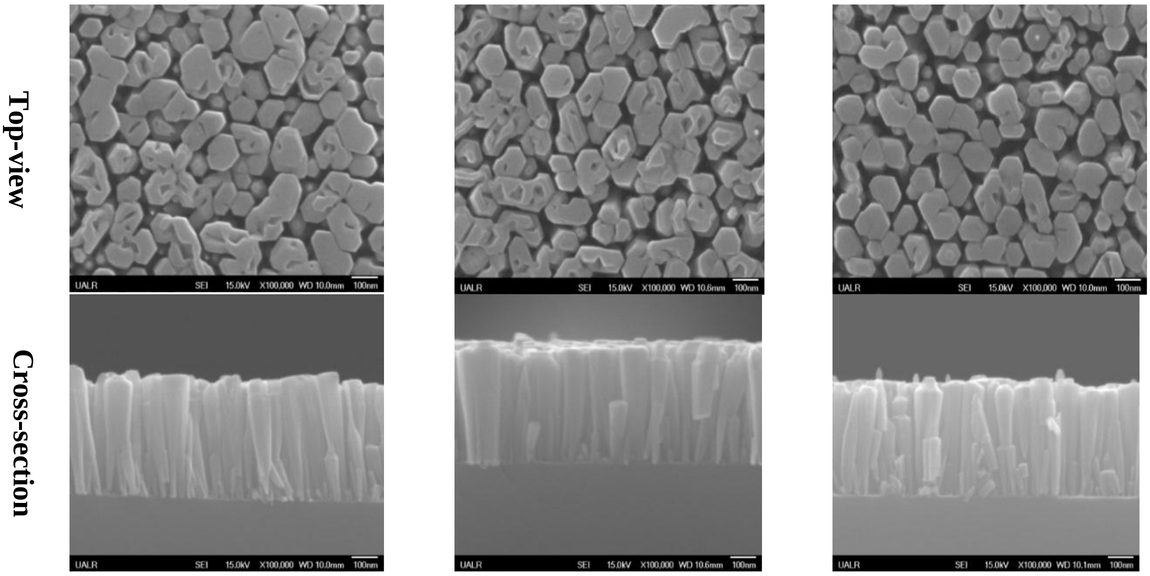
The InGaN samples were grown under N-rich condition with a constant beam equivalent pressure (BEP) of Gallium, Indium and Nitrogen flux, only plasma power has been changed. The detail of the growth parameters for InGaN samples is summarized in Table 1. The growth temperature and growth time were kept at 575°C and 90 minutes respectively. The top and cross section view of InGaN samples was characterized by using scanning electron microscope (SEM) measurement. Besides, the optical properties and the In composition of the samples were investigated by room temperature photoluminescence (PL) measurement, at which the samples were excited by 150 W white-light source of Xe lamp, monochromatically filtered to give 0.1 W/cm^2 power density at λ = 320 nm.

Table 1: Growth parameters and conditions for InGaN nanorods grown by PAMBE

Sample number	FGa/FN	FIn/FN	Plasma power (W)
1	0.54×10^{-2}	0.80×10^{-2}	325
2	0.54×10^{-2}	0.74×10^{-2}	375
3	0.54×10^{-2}	0.73×10^{-2}	400

3. Results and Discussion

The set of InGaN (0001) samples were grown on Si (111) substrate with different plasma power at 325, 375, and 400 W as shown in the Figure 1. From the SEM images, the average density, diameter, and height of InGaN nanorods were 4.8×10^{9} cm^{-2}, 71.3 nm, and 398 nm respectively for the lowest plasma power, while they were 2.5×10^{9} cm^{-2}, 107.6 nm and 445 nm respectively with increasing plasma to 400 W. Note that the non-uniformity in the shape and size expected to be originated from the nucleation stage in the sense of that the nucleation does not occur simultaneously, giving a broad dispersion in heights and sizes (Ristić *et al.*, 2008).



Sample 1 **Sample 2** **Sample 3**

Figure 1: Top-view and cross-section SEM images of InGaN samples as a function of nitrogen plasma power

The room temperature photoluminescence (RT-PL) spectra for the samples were measured as shown in the Figure 2. It was seen with increasing plasma power from 325 to 400 W that the PL bands were shifted from 2.33 eV to 2.21 eV, as a result of increasing indium concentration from 31.4% to 33.7% by using Vegard's law (Neugebauer *et al.*, 1995). The crystal quality of InGaN was further investigated by using the full width at half maximum (FWHM) of the PL peaks. The FWHM values of PL peaks from InGaN samples for the lowest and highest plasma power were measured which were 0.273 eV and 0.281 eV respectively. Indicating that, with increasing In content the crystal quality was further degraded due to the random alloying distribution between InN and GaN. Note that a single peak of the PL emission implies the absence of the phase separation, the suppressed phase separation under N-rich condition in general can be explained due to reducing of the surface migration of group-III adatoms under excess nitrogen-rich condition (Komaki, Katayama, Onabe, Ozeki, & Ikari, 2007).

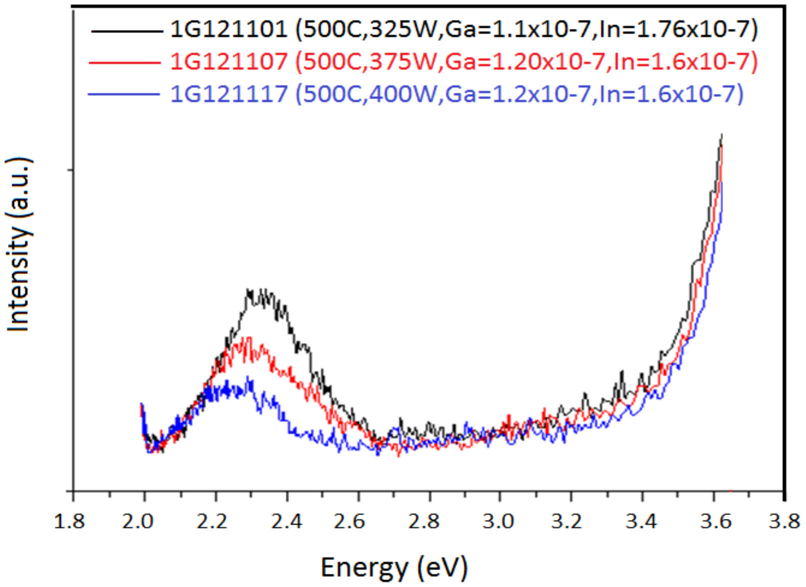


Figure 2: Photoluminescence spectrum from InGaN samples for different plasma power

The electronic and vibration of excited states of nitrogen molecules are plotted in the Figure 3, it consisted of number of broadband corresponding to first positive from $C^3\Pi_u$ to $B^3\Pi_g$ and second positive from $B^3\Pi_g$ to $A^3\Sigma_u^+$ of excited nitrogen molecules emission as well as some of sharp lines corresponding to excited atomic nitrogen from $3p^4S^0$, $3p^4P^0$, $3p^4D^0$. In addition to ionized atomic or molecules nitrogen species (Hughes *et al.*, 1995; Kushi *et al.*, 1999; Vaudo, Cook, & Schetzina, 1994), it is known that the excited nitrogen species within plasma cell have positive effect on crystal growth but occasionally they have negative effect on the growth depending on the energy delivery of the excited nitrogen species to the surface structure (Jordan, Burns, & Doak, 2001).

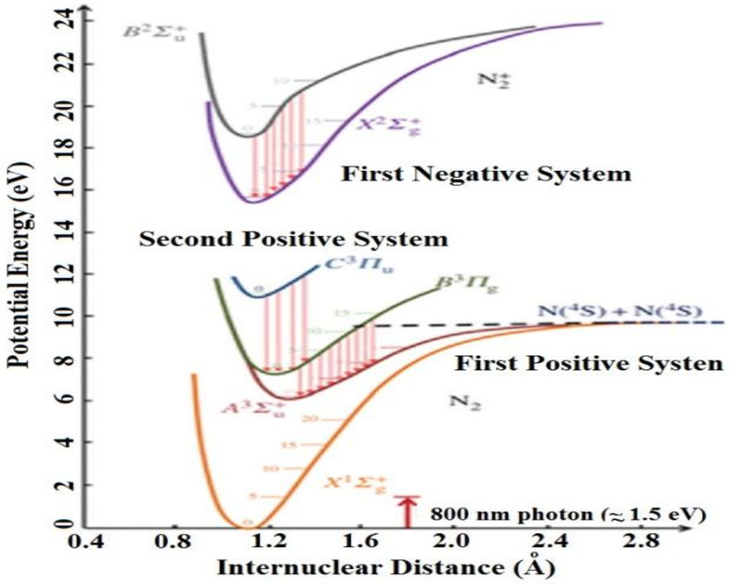


Figure 3: Electronic and vibration levels of excited species from nitrogen plasma (Morkoç, 2009)

The integrated intensities of excited atomic and molecular nitrogen species in the plasma cell were quantified for the different plasma powers as shown in the Figure 4. The emission peak intensity of excited atomic nitrogen rises with the RF plasma power, while the intensity of first and second positive of excited nitrogen molecular was totally different, and it is nearly constant with altering RF plasma power. A very small increasing of peak intensity of nitrogen molecules with power is observed in the case of RF power up to 400 W (Iliopoulos *et al.*, 2005; Kim, Lee, Park, & Park, 2000). Also, the peak intensity originated from the nitrogen ions is negligible due to low intensity of the emission spectrum which is not shown here. Since the atomic nitrogen transition can be produced within plasma cell, so at a certain level of nitrogen flow rate 2.2×10^{-5} Torr, with increasing plasma power from 325 to 400 W the atomic intensity has been increased. This observation can be explained by increasing the cracking efficiency of nitrogen molecules within RF plasma cell. Since there is a sufficient energy to dissociate all nitrogen molecules to nitrogen atoms at higher plasma power, thus the intensity of atomic nitrogen species which is proportional to the population density was increased (Ptak, 2001).

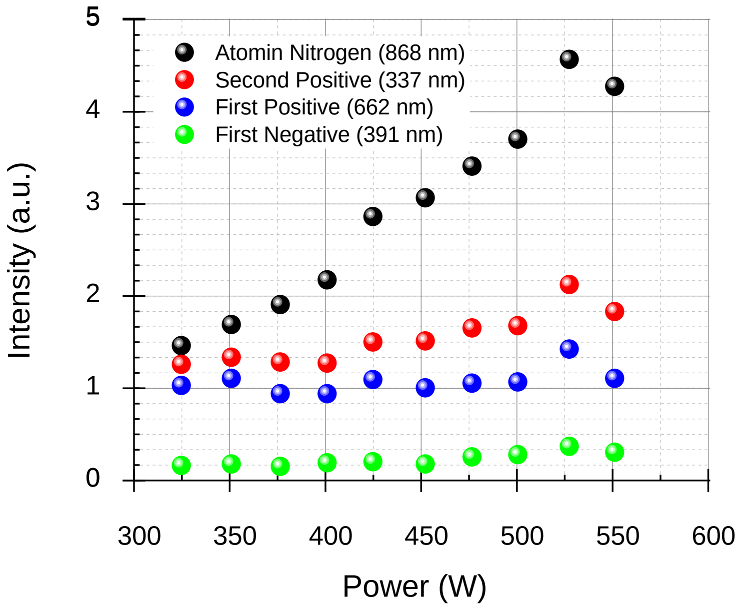


Figure 4: Nitrogen plasma species at different plasma power

Plasma power is one of the main growth parameters that affects the crystal quality and growth rate of InGaN structure. To demonstrate the growth rate of InGaN nanorods as a function of plasma, we study the energy delivery to InGaN nanorods by nitrogen plasma species as shown in the Figure 4.

From the Cross-section measurements of the samples by SEM as presented in the Figure 1, we observed the growth rate of the InGaN samples was increased gradually from 253 to 262 nm/h by rising plasma power from 325 to 400 W, as revealed in the Figure 5, subsequently an increase of the intensity of the excited atomic nitrogen emission was remarked, while the intensity of nitrogen molecular species was almost independent on the plasma power. Thus, it is concluded that the excited atomic nitrogen is the most active form among other species participating the growth of InGaN materials. The independent of InGaN growth on the intensity of molecular nitrogen species is a good indicator of cracking efficiency decreasing the plasma cavity at higher plasma power. These results are in a good agreement with observations work done by Osaka *et al.* (2007) and Kim *et al.* (2000).

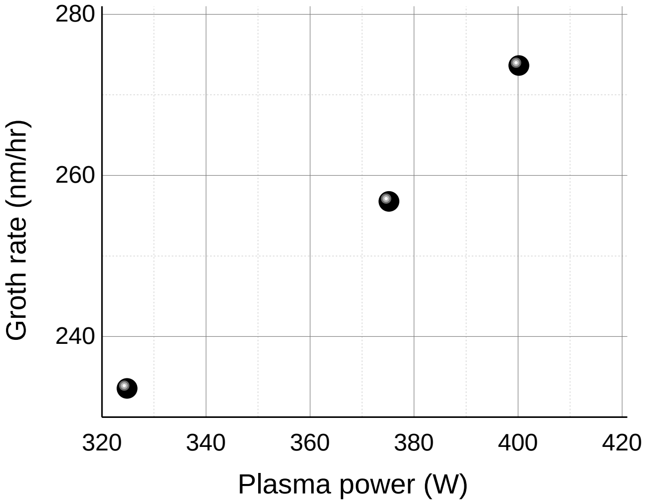


Figure 5: Growth rate of InGaN as a function of plasma power

4. Conclusions

In this study, we focused on the performance of the excited atomic and molecular nitrogen flux for enhance III-nitrides growth by PA-MBE. This was qualitatively studied through optical emission spectroscopy versus RF plasma power. The results showed that the excited atomic nitrogen was elevated with plasma power while the second positive nitrogen molecules stayed independently. Thus, plasma power can be tuned to operate the optimal conditions to control the quantity of excited nitrogen species which exist in the plasma cavity. Finally, the atomic flux was primarily responsible for growth of InGaN nanorods.

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