Molecular beam epitaxy of N-polar InGaN

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We report on the growth of N-polar $In_xGa_{1-x}N$ by N_2 plasma-assisted molecular beam epitaxy. Ga-polar and N-polar InGaN films were grown at different growth temperatures and the composition was estimated by photoluminescence (PL) measurements. A growth model that incorporates the incoming and desorbing atomic fluxes is proposed to explain the compositional dependence of InGaN on the flux of incoming atomic species and growth temperature. The growth model is found to be in agreement with the experimental data. The peak PL intensity for N-face samples is found to exhibit a two order of magnitude increase for a 100 °C increase in growth temperature. Besides, at 600 nm, the N-face sample shows more than 100 times higher PL intensity than Ga-face sample at comparable wavelengths indicating its superior optical quality. The understanding of growth kinetics of InGaN presented here will guide the growth of N-polar InGaN in a wide range of growth temperatures. © *2010 American Institute of Physics*.
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The InGaN alloy system has attracted much research over the years due to its wide applications in optoelectronic devices, especially for achieving wavelengths[1,2] which are either very difficult or impossible with established As–P based technology. High power white light sources and blue lasers based on InGaN/GaN technology are now commercially available. However, green lasers with high power and efficiency have not been reported yet.[3] The availability of InGaN with band gaps in the green optical energy range is very promising for both green light emitting diodes and laser diodes. InGaN has also attracted interest for possible multijunction solar cells[4,5] since it can access almost the entire solar spectrum for maximum efficiency.[6]

However, the growth of high quality InGaN especially with the higher In-composition needed for green emission still poses severe challenges. This is due to the fact that optimal growth conditions for the alloy components InN and GaN are significantly different. Growth of conventional Ga-polar InGaN by plasma-assisted molecular beam epitaxy (PAMBE) is limited to temperatures lower than 500 °C (Ref. 7) as InN decomposes at temperatures higher than this. Since this is significantly lower than optimal growth temperatures needed for growth of GaN, the quality of InGaN films is compromised. More recently, PAMBE grown InGaN based light-emitting diodes (LEDs) have been shown to exhibit output power of 9 mW at 385–400 nm[8] and 3 mW at 450 nm (Ref. 9) at injection currents of 1 A and 0.1 A, respectively, which however are still far from touching the green or 520 nm mark.

Recently, the reversed direction of polarization of GaN, i.e., N-polar orientation was explored to exploit advantages for high-speed performance of highly scaled transistors.[10,11] It is also shown that PAMBE growth of N-face InN can be done at 100 °C higher than the thermal dissociation limit of In-face InN.[12–15] This opens up the possibility of growing N-face InGaN with higher In-compositions at higher growth temperatures than is possible with In(Ga)-face InGaN. However, few reports on N-face InGaN growth have been published so far.[16,17] This report aims to provide an understanding of indium incorporation in N-face InGaN as compared to Ga-face InGaN with respect to change in growth temperatures. The In-composition as well as the optical quality of the InGaN films is assessed by room temperature photoluminescence (PL) measurements.

Samples used in this study were grown by PAMBE in a Veeco Gen 930 system equipped with standard effusion cells for Ga and In. Active nitrogen was supplied using a Veeco rf plasma source. The substrates used were N-face free standing LED quality GaN templates (dislocation density $\sim 10^8$ cm^{-2}) obtained from Lumilog[18] and Ga-face GaN on sapphire template (dislocation density $\sim 10^9$ cm^{-2}) from Kyma.[19] Both N-face and Ga-face templates were coloaded on a single silicon wafer using indium-bonding to ensure identical growth conditions so that the difference in In-incorporation for InGaN of both polarities at a particular growth temperature could be studied. Since sapphire has a lower thermal conductivity than GaN, the surface temperature (using optical pyrometer) of the Ga-polar sample was found to be 3–5 °C higher than that of the N-polar sample. Even with this taken into account, Indium incorporation in the N-polar sample is, as expected, significantly higher than the Ga-polar sample. All InGaN layers discussed in this work were grown for 1 h with a standard GaN growth rate of 5 nm/min; however due to decomposition of InGaN, the actual thickness of the films is expected to be less than 300 nm.

Three sets of growths were done at growth temperatures of 500, 550, and 600 °C in an In-rich regime, the temperatures being stabilized prior to growth. The growth temperature was monitored by an optical pyrometer with readings calibrated against the melting point of Al. Since In-incorporation depends on growth temperature as well as on Ga/N flux ratio,[10] the Ga-flux was kept constant at 9.5×10^{-8} Torr. An rf power of 350 W and N_2 flow rate

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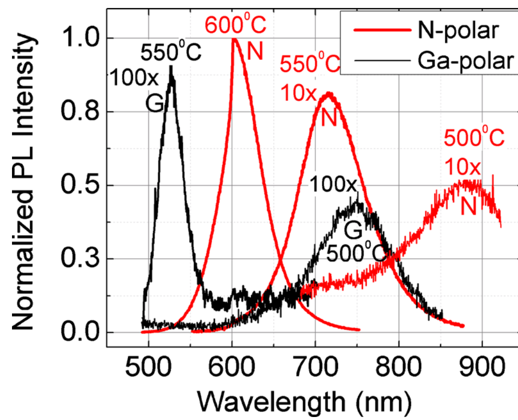

FIG. 1. (Color online) PL intensity (normalized with respect to the highest intensity obtained) of Ga-face and N-face InGaN on a linear scale; the magnification factor, polarity (G for Ga-polar and N for N-polar), and growth temperature are indicated against each peak.

equivalent to 1.9×10^{-5} Torr of chamber pressure was used throughout.

Light emission from the N-face and Ga-face InGaN samples was collected by 1 m long monochromator with a ruled grating 1200 gr/mm placed at surface normal and the signals were detected by Hamamatsu R2658 photomultiplier tube mounted exit slit of the monochromator. A Stanford Research System SR830 DSP lock-in amplifier was used to analyze the data. Ar laser with a wavelength of 488 nm was used as an excitation light source. A control sample was used to confirm that the measurements were performed in identical conditions so that PL intensity values between each measurement performed can be comparable. All measurements were done at room temperature.

Figure 1 shows the PL intensity of samples grown at different temperatures for both polarities on a linear scale. To bring the values to comparable intensity level, the intensities of Ga-face samples have been magnified 100× while those of the N-face samples (except for the one at 600 nm) have been magnified 10×. The sharpness of the peak and its high intensity for the N-face sample at 600 nm compared to other samples is thus well discernible.

Figure 2 shows on its left axis the peak intensity corresponding to the PL measurements of each sample against the corresponding wavelength while full width at half-maximum (FWHM) of the same have been shown on the right axis. The peak intensity for N-face samples show more than two orders

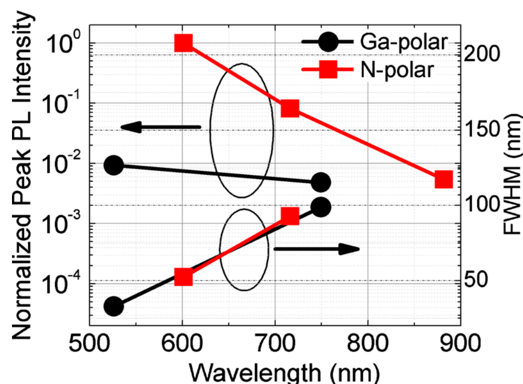

FIG. 2. (Color online) Peak PL intensity (normalized with respect to the highest intensity obtained) and FWHM of InGaN samples of both polarities as a function of wavelengths (corresponding to band gaps extracted from room temperature PL measurements). The lines are a guide to the eyes.

TABLE I. Energy band-gap values extracted from PL measurements as well as In mole-fractions deduced using bowing parameters (Ref. 22) for InGaN of both polarities.

Growth temperature (°C)	Band gap (from PL) (eV)		In mole-fraction (%)	
	Ga-face InGaN	N-face InGaN	Ga-face InGaN	N-face InGaN
500	1.655	1.406	43.6	51.1
550	2.357	1.731	24.6	41
600	>2.541	2.062	<20	32.2

of magnitude increase as wavelength decreases from ~900 to ~600 nm corresponding to a 100 °C increase in growth temperature. Besides, the N-face sample shows much higher intensity than the Ga-face sample, especially at shorter wavelengths the N-face sample has a two order of magnitude higher intensity. This holds promise for achieving superior luminescence property of N-face InGaN sample as the wavelength is reduced further. The FWHM decreases at lower wavelengths which further indicate better optical quality at higher growth temperatures. Our observations are in contrast to the reports by Masui *et al.*[20] where they observe better optical and luminescence qualities for metal-organic chemical vapor deposition grown Ga-face InGaN multiquantum well (MQW) than N-face InGaN MQW as well as to the reports by Shen *et al.*[21] where they claim superior optical qualities for PAMBE grown Ga-face InGaN over N-face samples.

The energy band-gap values extracted from the room temperature PL peaks are tabulated in Table I. The corresponding In-compositions in the samples were deduced using bowing parameters from Ref. 22. The wavelength corresponding to the Ga-face InGaN grown at 600 °C was out of the range of the PL setup, that is, its wavelength due to its very low In-composition, was lower than the excitation light source wavelength (488 nm, >2.541 eV). Thus it has been ignored in this study. $\omega-2\theta$ triple axis scans of the N-polar films performed using a BEDE high resolution x-ray diffraction (XRD) system showed single peaks, and no phase segregation or compositional nonuniformity. The In composition from the XRD data agrees with the PL data, if we assume that the films are partially relaxed. The important observations from Table I are first, as expected, the In-composition and hence In-incorporation drops as growth temperature increases due to higher decomposition of InN at higher temperatures irrespective of the polarity. Second, for a given growth temperature, the In-incorporation is higher for N-face InGaN than for Ga-face InGaN. This implies that N-face InGaN can be grown at a higher temperature for a specified In-mole fraction than Ga-face InGaN.

We propose a growth model for N-face InGaN based on InN decomposition which explains the dependence of In-composition on growth temperature. The model has been developed for the metal rich N-limited growth regime where the active N-flux available determines the growth rate with an excess of In on the surface. It has been shown[15] that N-polar InN decomposition can be considered to be as N-atoms leaving the surface, which we model schematically in the inset of Fig. 3 as N-atoms leaving the surface. The decomposition rate F^D will hence be proportional to the In-mole fraction "x" in the $In_xGa_{1-x}N$ film (and thus for a pure GaN film, there will be no decomposition as expected). It has

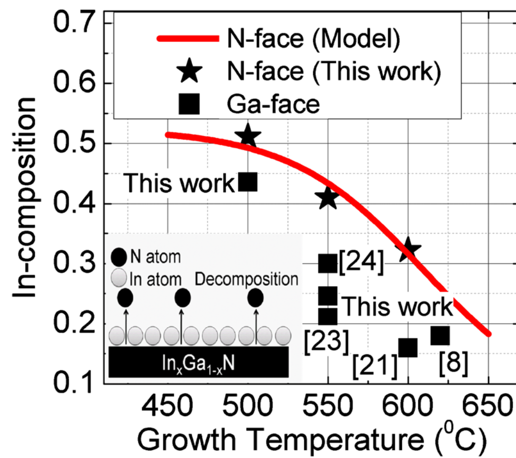

FIG. 3. (Color online) Variation in In-composition with growth temperature for N-face InGaN obtained in this study along with the curve from our growth model. A few data points corresponding to Ga-face InGaN reported elsewhere are included to establish the achievability of higher In-incorporation for N-face polarity. Inset: Schematic showing the modeling of InN decomposition as N-atoms leaving the surface.

also been established[15] that in PAMBE growth of N-polar InN, metallic In accumulation (adlayer and droplets) on the surface due to In flux from InN decomposition and impinging In atoms from the source is limited by maximum In-desorption rate F^{des} up to a growth temperature of ~610 °C. However, at higher growth temperatures above ~610 °C, desorption rate of In exceeds decomposition rate of InN. Thus excess In can remain on the surface in the form of droplets only if the impinging In-flux is greater than $|F^{des}-F^{D}|$. Else if the surface is devoid of excess In and there is an excess of N, the growth will shift to In-limited regime instead of being N-limited and our model will be invalid. Since all our growths have been performed in an In-rich regime at temperatures below ~610 °C, the In-incorporation will depend only on the reduced N-flux F^{N*} and the Ga-flux F^{Ga} which remains unchanged. The Ga-flux to achieve an In-composition of "x" in an $In_xGa_{1-x}N$ film can be hence written as $F^{Ga}=(1-x)F^{N^*}$, or the In-composition can be written as $x=1-(F^{Ga}/F^{N*})$. The decomposition rate F^{D} is also shown to have an Arrhenius dependence on substrate temperature T_{sub} (Ref. 15) as $F^{D}\propto x\exp(-E_a/kT)$, where E_a is the activation energy of decomposition of N-face InN which has been calculated earlier to be 1.2 eV.[15] The reduced nitrogen flux which determines the growth rate now can be written as $F^{N*}=F^{N}-F^{D}$ where F^{N} is the N-flux in the absence of any decomposition. Using this set of equations, the In-composition in the InGaN film can be expressed as a function of temperature.

Using these sets of equations and the data points of In-composition corresponding to the three N-face InGaN samples, a curve fitting was done to evaluate the proportionality factor in the equation relating decomposition rate to the temperature. The resulting variation in "x" as a function of substrate temperature as obtained from our model is plotted in Fig. 3. The data points are found to exhibit a very good fit with the curve validating our growth model.

Several data points showing In composition and substrate temperature for Ga-face InGaN by MBE from the literature[8,21,23,24] have been included in Fig. 3 as a comparison. The advantages of using the N-polar orientation are obvious from the plot. For any Indium composition, the N-polar orientation allows for higher growth temperatures than Ga-polar. As the In composition is increased, the difference becomes more pronounced.

In conclusion, a temperature dependent comparative growth study of N-face and Ga-face InGaN grown by plasma assisted MBE was done. Based on the band gaps and hence corresponding wavelengths as determined by room temperature PL measurements, it was found the indium incorporation efficiency in N-face InGaN was higher than in Ga-face InGaN at a given substrate temperature. More importantly, N-face InGaN sample shows significantly higher (100× times) peak PL intensity than Ga-face sample particularly at shorter wavelengths. A growth model was proposed to explain quantitatively the variation in In-composition as a function of growth temperature based on InN decomposition. The calculated curve was found to fit the experimental data points well.

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