Comprehensive study of the optical and physical properties of InGaN nanocolumns grown on Si(111) and por-Si/Si(111) substrates by plasma-assisted molecular beam epitaxy

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Abstract. We report on a successful growth of self-organized In$_{0.3}$Ga$_{0.7}$N nanorods by low-temperature plasma-assisted molecular beam epitaxy on a Si(111) substrate with and without preformed thin porous Si layer (por-Si). XPS study of both samples confirmed In content predicted according to the growth conditions as well as the analysis of PL spectra. Moreover, InGaN nanorods grown on por-Si layer showed ~20% higher PL intensity.

1. Introduction

III-nitride semiconductors are of great interest for applications in high temperature/power electronic devices as well as photonic devices in a wide spectral range from ultraviolet (the GaN bandgap is 3.45 eV [1]) to near-infrared (the InN bandgap is ~0.7 eV [2]). However, due to the lack of the native GaN substrates, III-N based structures conventionally grow on heteroepitaxial substrates with a high thermal expansion coefficient and lattice mismatches leading to high threading dislocation generation. Many dislocation reduction techniques such as lateral overgrowth, low-temperature buffer layer, multiple intermediate layers have been already developed. However it is still difficult to reduce the dislocation density to be less than $10^6$ cm$^{-2}$. It is well known that large nanorod free surface helps to suppress propagation of the threading dislocations and stress generation [3], thus InGaN nanorods structure seems to be suitable for fabrication light-emitting devices with a high efficiency.

In this paper we report on fabrication and comprehensive study of the InGaN nanorods structure grown on a compliant Si-substrate by a low temperature plasma-assisted MBE (LT PA MBE) with up to 20% higher PL intensity than ones formed on traditional Si(111) substrate.

2. Experimental

All Si substrates were pre-treated using the Shiraki method [4]. Then a 20 nm-thick por-Si layer was formed on several substrates using the original method of selective etching. After that all the substrates was annealed at 820°C for an hour until the 7x7 surface reconstruction was observed by RHEED. All of the growth processes were performed by using Veeco Gen 200 PA MBE setup. For comparison nanorods were grown in the one growth process on conventional Si(111) substrate and substrate with a por-Si layer (sample a and b, respectively) without nitridation step. LT InGaN nanorods were formed at $F_{III}/F_N$=1 flux ratio, whereas fluxes was $F_{Ga}$=0.04 ML/s, $F_{In}$=0.02 ML/s and $F_N$=0.06 ML/s and the
substrate temperature of $T_s = 400^\circ C$. RHEED was used for *in situ* control of the surface morphology of the layers. The surface morphology also was investigated *ex situ* by atomic force microscope (AFM), scanning electron microscopy (SEM) and with optical microscope (OM). Crystalline quality was studied by XRD analysis, mobility and carrier concentration was determined by analysis of the Hall measurements data. Ternary alloy composition was confirmed by XPS and PL spectra analysis.

3. Results

The lateral sizes distribution histogram obtained from the AFM study of the substrate and samples surfaces (Figure 1) shows that the use of a por-Si layer has a crucial influence on the distribution of the nanorods diameters.

![Figure 1](image1.png)

**Figure 1.** The lateral sizes distribution histogram obtained from AFM study of por-Si layer (left), InGaN/Si(111) layer (centre) and InGaN/por-Si/Si(111) layer (right).

In addition, cross section SEM images of all heterostructures (Figure 2) illustrate rather planar morphology for all of the interfaces. One can also observe a more pronounced nanocolumnar structure in case of the sample b.

![Figure 2](image2.png)

**Figure 2** Cross-section SEM images of the GaN/Si(111) (left) and GaN/por-Si/Si (right) nanorods.

XPS spectra of both samples are listed at figure 3.
Figure 3 XPS spectra of the InGaN/Si(111) (a) and InGaN/por-Si/Si (b) nanorods.

PL spectra of the samples which are listed at Figure 4 revealed ~20% higher PL peak intensity for sample grown on por-Si layer.

Figure 4 PL spectra of the InGaN/Si(111) (a) and InGaN/por-Si/Si (b) nanorods.
**4. Discussion**

The threading dislocations reduce the emission efficiency, increase the leakage current, and reduce the life time of III-N based devices. Usage of nanorods-like structure is the one of the possible ways to create energy efficient LED structures with high light extraction surface due to presence of the large free nanorods surface, which acts as dislocation suppression instrument due to their inclination. AFM and SEM study of the surface morphology confirmed nanorods-like structure of the samples and increased average distance between them. Thus, since the average distance between nanorods was increased – light extraction coefficient should become higher. This fact can be confirmed by PL spectra analysis of both samples. As can be seen from figure 4 the maximum PL intensity of sample b grown on *por-Si* layer is by ~20% higher than that one for sample a grown on Si(111) substrate. Also PL maximum peaks intensity positions are 585 nm and 574 nm for sample a and b, respectively. We believe that this peak position shift is related with ternary alloy composition fluctuation as well as with the difference in the elastic strain in the InGaN layers. As it was shown in K. O’Donnell work [5] PL peak position is related indirectly with InGaN bandgap and it can be evaluated as

\[
E_{pl}(x) = -1.54 + 1.45E_g(x) \tag{1}
\]

The \(E_g(x)\) dependence for In\(_{x}\)Ga\(_{1-x}\)N alloys

\[
E_g(x) = 3.51 - 2.75x \tag{2}
\]

Using equations (3) and (4) we can calculate In composition in In\(_{x}\)Ga\(_{1-x}\)N ternary alloy as \(x_a \approx x_b \approx 0.33-0.34\) which correlates well with growth conditions. Moreover, as it was shown in [6] In content can be evaluated from XPS spectra of the samples as

\[
X_h = \frac{I_{h_{4d5}}/F_{h_{4d5}}}{I_{h_{4d5}}/F_{h_{4d5}} + I_{Ga_{2p3}}/F_{Ga_{2p3}}} \tag{3}
\]

Whereas \(I_i\) – integrated peak intensity and subscript denotes peak type and \(F\) – correlation coefficient (\(F_{Ga_{2p3}}=2.75\) and \(F_{h_{4d5}}=4.53\)). Using formula (3) we can calculate In content as \(x_a = 0.34\) and \(x_b = 0.32\). It should be noted that basing on XPS analysis we can assume that the free surface of nanorods is oxidized, that is confirmed by asymmetrical form of the In\(_3\)d\(_5\) peaks.

As can be seen from the data obtained from XRD analysis (table 1) In\(_{x}\)Ga\(_{1-x}\)N layer grown on *por-Si* layer have slightly better crystalline quality.

The van der Pauw–Hall-effect measurements were performed at room temperature. Both samples revealed p-type conductivity with carrier concentration of \(3 \times 10^{19}\) cm\(^{-3}\) and \(3.64 \times 10^{19}\) cm\(^{-3}\) and carrier mobility of 104.7 cm\(^2\)/(V·s) and 103.4 cm\(^2\)/(V·s) for samples a and b, respectively.

Summarizing this discussion, InGaN nanorod-like layers with reasonable crystalline quality and p-type conductivity were formed by plasma-assisted molecular beam epitaxy on compliant *por-Si* and traditional Si(111) substrates. PL and XPS study independently confirmed \(x \approx 0.32-0.34\) In content in both samples. PL spectra also revealed up to 20% higher peak intensity growth for InGaN layer formed on *por-Si* layer. AFM measurement of nanorods diameter distribution showed that more than 75% of nanorods grown on the compliant substrate have the same diameters ~ 40 nm contrary to nanorods grown on Si(111) substrates which diameters are statistically distributed in range of 20–60 nm. Thus, usage of compliant Si substrates with nano-porous Si layer seem to be a suitable way to integrate III-N technology with existing Si technology.

### Table 1. Data obtained from XRD analysis

| Sample | c, Å | a, Å | \(e_{xx}\) | \(\varepsilon_{zz}\) | \(\rho_{core}\), cm\(^{-1}\) | \(\rho_{edge}\), cm\(^{-1}\) |
|--------|------|------|-----------|----------------|-----------------|-----------------|
| a      | 5.4005 | 3.0178 | -0.087 | 7.06 \times 10\(^{-3}\) | 3.15 \times 10\(^{10}\) | 1.97 \times 10\(^{8}\) |
| b      | 5.4156 | 3.0253 | -0.086 | 7.04 \times 10\(^{-3}\) | 3.10 \times 10\(^{10}\) | 1.60 \times 10\(^{9}\) |
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