

**Polar-Plane-Free Faceted InGaN-LEDs
toward Highly Radiative Polychromatic Emitters**

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2020

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Abstract

III-nitride semiconductors (Al, Ga, In)N offer a wide direct bandgap that can be flexibly tuned over the complete spectral range from deep ultraviolet (~ 6.2 eV) to near infrared (~ 0.7 eV). This means the theoretical possibilities of arbitral spectral syntheses in the entire visible spectral range based on III-nitride systems. In addition, nonpolar and semipolar crystallographic planes of III-nitrides are expected to show superior optical transition probabilities due to the weaker internal electric fields in the InGaN active layers, compared to the conventional polar III-nitrides. Therefore, if the bandgap flexibility and semi/nonpolar physics can be well combined in some novel ways, that may lead to highly efficient and color-tunable light emitters. In this study, aiming at exploring such novel optoelectronics, InGaN-based three-dimensional (3D) structures grown on semipolar GaN substrates by selective area growth technique were investigated. The spontaneously-formed 3D structures induce microscopically spatial distribution of InGaN alloy composition and thickness, leading to the polychromatic emission properties without phosphors. We selected GaN substrates for homoepitaxy because the heteroepitaxially grown semipolar films usually suffer from numerous defects. As the substrate orientations, semipolar $\{11\bar{2}2\}$ plane was chosen, because the improved optical transition probabilities were theoretically and experimentally demonstrated.

To explore microstructures on semipolar $\{11\bar{2}2\}$ plane, 3D faceted GaN structures were grown on both $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ GaN substrates with various stripe mask geometries. It was found that $[\bar{1}\bar{1}23]$ stripe directions with particular mask designs on both $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ surfaces provide polar-plane-free faceted 3D structures, composed of semipolar $\{11\bar{2}2\}$ and $\{1\bar{1}01\}$ and nonpolar $\{1\bar{1}00\}$ facets. The growth processes of the 3D GaN structures on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ were observed by AlGaN marker technique and were found to be quite different. The observed growth evaluations suggest that the distinct shape evolutions between the 3D GaN structures on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ are well accounted for by the inter-facet surface diffusion, which could be affected by the surface

bond configurations of the crystallographic orientations forming the 3D structures.

InGaN QWs were grown on the developed polar-plane-free structures on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ in order to investigate the emission characteristics. It was found that the 3D InGaN QWs on $(\bar{1}\bar{1}2\bar{2})$ exhibit facet-dependent polychromatic emission properties, whereas those on $(11\bar{2}2)$ showed monochromatic emission. These emission properties of the 3D InGaN QWs on $(\bar{1}\bar{1}2\bar{2})$ and $(11\bar{2}2)$ were attributed to the inter-facet variations of the QW structural properties, which are derived from the inter-facet In adatom migrations affected by the crystallographic orientation dependence of In incorporation. Finally, quite short radiative lifetimes from all the facet QWs were experimentally demonstrated for the 3D InGaN QWs on $(\bar{1}\bar{1}2\bar{2})$. Hereafter, efforts in this study were directed to the polar-plane-free faceted 3D structures on $(\bar{1}\bar{1}2\bar{2})$.

To realize polar-plane-free faceted 3D LEDs, firstly, growth conditions for p-type and n-type doping of $(\bar{1}\bar{1}2\bar{2})$ planar LEDs were investigated. Particularly, Mg doping conditions for p-type GaN strongly depend on crystallographic orientations, and the conventional doping condition for $(11\bar{2}2)$ planar LEDs was found to be unfavorable for the $(\bar{1}\bar{1}2\bar{2})$ plane. As the mechanism behind it, dopant profile broadening of intentional impurities and higher concentrations of unintentional impurities degraded the device performance. Thus, the doping conditions were optimized by adjusting growth pressures and Mg/Ga flow ratios, and the $(\bar{1}\bar{1}2\bar{2})$ LED with bright electroluminescence (EL) was obtained. Finally, the developed growth condition for $(\bar{1}\bar{1}2\bar{2})$ planar LEDs was applied to the growth of polar-plane-free faceted 3D LED structures. A SEM observation indicated successful pn-junction formation on all the facet in the 3D structures.

Finally, all the above results were summarized in electrically driven, polar-plane-free faceted 3D LEDs on $(\bar{1}\bar{1}2\bar{2})$. After establishing an LED device processing to the 3D LED structures, the device performance was evaluated by electrically injected conditions at room temperature. The 3D LEDs exhibited rectifying properties, though the turn-on voltages and series resistances were higher than those of the $(\bar{1}\bar{1}2\bar{2})$ planar LEDs, probably due to the premature device processes. Moreover, EL spectrum from the 3D LEDs was well controlled by altering the mask geometry. Although the emission intensity was quite low, whitish color was also synthesized by the 3D LEDs. In any case, the obtained results indicate the possibility of realizing highly efficient polychromatic LEDs based on 3D faceted InGaN LEDs, excluding the polarization issue derived from the (0001) polar plane.

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Chapter 1

Introduction

III-nitride semiconductors (Al, Ga, In)N have opened a new era of visible and ultra-violet (UV) light emitters. For the visible region, where is the subject of this study, InGaN-based light-emitting diodes (LEDs) and laser diodes (LDs) already arise a significant technological impact. Blue and green LEDs, white LEDs composed of blue LEDs and yellow phosphors, and blue and green LDs are implemented in commercially available products. A part of these achievements can be attributed to the unique material properties of the InGaN system, offering a wide direct bandgap that can be flexibly tuned over the visible spectral region [0.7 eV (InN) to 3.5 eV (GaN)] depending on the In composition [1, 2]. Here, reconsidering the fundamental physical properties, the wide bandgap tunability suggests arbitral visible spectra synthesis based on the InGaN systems in some ways. Multi-chip semiconductor light emitters have been proposed to mix the light of different colors emitted by a few primary LED chips [3], as schematically shown in Fig. 1.1(a) and 1.1(b). The structure is composed of red-to-yellow AlGaInP LEDs and blue-green InGaN LEDs integrated by the device process technology through the *top-down approach*. This kind of LED design should be superior in efficiency and longevity to a conventional phosphor-based polychromatic LED and also allows for color adjustment by varying the contribution of emissions from the primary LEDs. Meanwhile, in the *bottom-up approach*, InGaN-based nano- and micro-structures based on advanced crystal growth techniques are emerging as platforms for monolithic polychromatic LEDs. Figure 1.1(b) and 1.1(c) show conceptual diagram of such an one-chip RGB LED and a luminescence spectrum, respectively. In this work, we shall note frontier of monolithically integrated polychromatic LEDs based on InGaN nano- and micro-structures.

As shown in Fig. 1.2(a), conventional monochromatic LEDs with InGaN systems

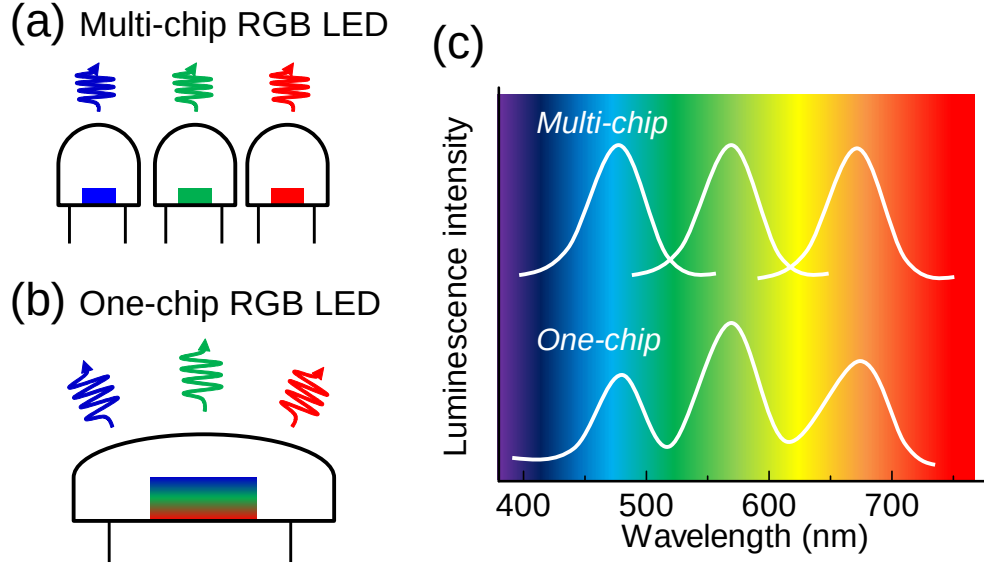


Figure 1.1: Schematics of (a) integrated RGB LEDs and (b) polychromatic LED on one chip, and (c) their luminescence spectra.

are grown on sapphire (0001) substrates and have planar structures. Thus, each stacked layer has a single uniform property, resulting in monochromatic emission. On the other hand, InGaN-based three-dimensional (3D) structures formed by selective-area epitaxial growth technique can be templates for monolithic polychromatic LEDs. Figure 1.2(a) depicts schematic of such a 3D LED structure. Regrowth of GaN on patterned silicon dioxide (SiO_2) masks by metalorganic vapor phase epitaxy (MOVPE) creates 3D structures, which consist of several facets, including (0001), $\{11\bar{2}2\}$, and $\{11\bar{2}0\}$. The three-dimensionality induces the spatial distribution of film properties of subsequently-grown InGaN active layers, which results in the facet-dependent polychromatic emissions. Funato *et al.* demonstrated proof-of-concept LEDs composed of 3D faceted InGaN LED structures [4, 5]. The polychromatic emission characteristics result in pastel color and white color syntheses. In addition, the higher light extraction efficiencies for the 3D LED structures than planar structures are predicted by simulations [4]. These impressive results motivated intensive research efforts toward phosphor-free polychromatic LEDs based on nitride 3D structures [4–16], and the findings show some promises of such 3D structures for future optoelectronics applications. Nevertheless, the challenges, including premature crystal growth and process difficulties, remain for practical applications. Before making clear the subject in this study, current issues in conventional InGaN-based LEDs with planar structures must be pointed out.

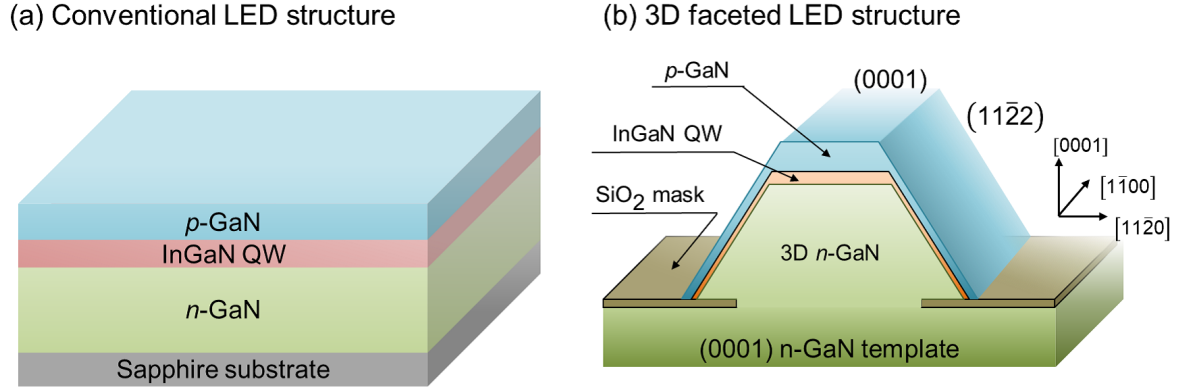


Figure 1.2: Conventional InGaN LED structure is planar, unlike (b) the 3D faceted InGaN LED structures.

As mentioned above, the wide-bandgap tunability of InGaN systems can cover the whole visible range depending on the In compositions. Very extensive works, however, have been dedicated to blue- and green-emitting InGaN LEDs. The research regarding longer wavelength emission corresponding to yellow or red region has been more restricted. Figure 1.3 presents reported external quantum efficiency (EQE) for commonly used InGaN LEDs on (0001) plane [17–22] and AlGaInP LEDs [23]. EQE gives a ratio of the number of useful photons to the number of injected electrons. EQE of InGaN LEDs drastically decreases toward the longer wavelength region, *i.e.*, In-rich InGaN active layers. This remarkable trend is essentially originated from distinct physical characteristics of InN and GaN. To illustrate the issues, figure 1.4 depicts a typical InGaN LED heterostructure together with the requirements for materials. Firstly, in terms of the thermodynamical nature, “phase separation” of InGaN alloys that becomes noticeable in In-rich InGaN layers significantly degrades the InGaN material quality, inducing numerous non-radiative centers. The large difference in interatomic spacing between InN and GaN gives rise to a solid phase miscibility gap. From another viewpoint, “stress” is a critical issue. The large lattice mismatch between InN and GaN ($\sim 11\%$) inevitably strains InGaN layers on GaN underlayers. If the stress is too large, lattice relaxation occurs through the formation of defects, which work as nonradiative recombination centers, to release the accumulated stress energy. The various approaches have been pursued to circumvent phase separation and stress issues by exploring crystal growth conditions or heterostructure designs. (For comprehensive reviews on In-rich InGaN heterostructures, see Ref. [24].) Finally, as the scope of interest in this study, “strain-induced piezoelectric fields” inside the active layers disturbing radiative recom-

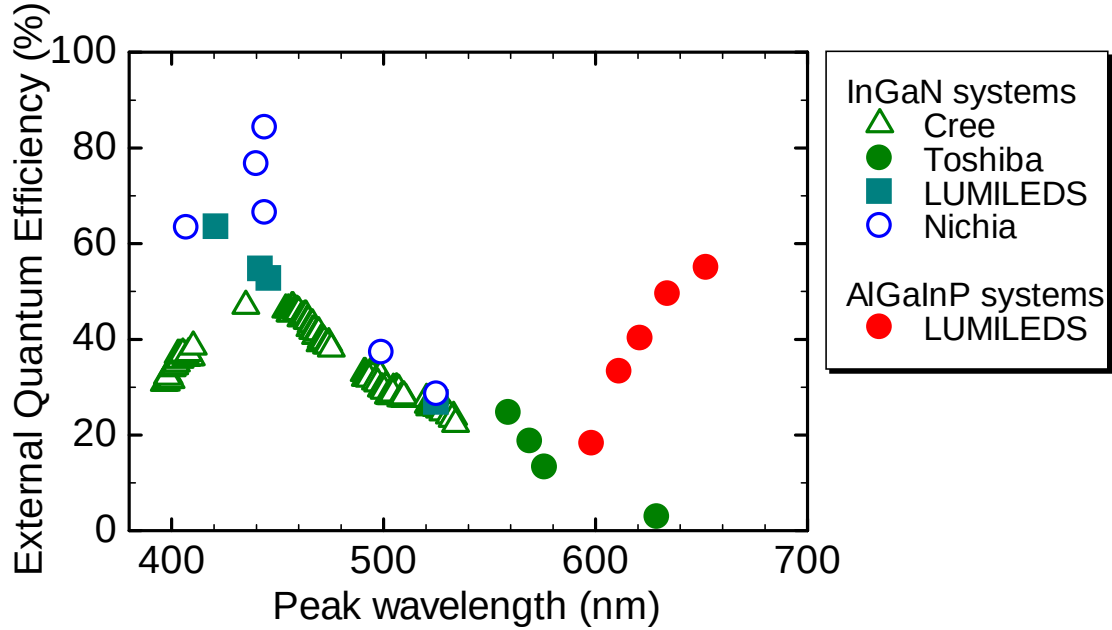


Figure 1.3: EQE as a function of emission wavelength for (0001) InGaN LEDs [17–22] and AlGaInP LEDs [23].

bination processes also become a crucial issue in the In-rich InGaN system. The details are described in the following.

Let us introduce the fundamentals of strain-induced piezoelectric fields and the recent progress of the attempts to solve this issue. A typical nitride semiconductor-based light emitter consists of *c*-oriented quantum wells (QWs), where the appreciable lattice mismatch between nitrides inevitably induces a piezoelectric polarization. This piezoelectric polarization, together with the spontaneous polarization, causes internal electric fields in the QWs, which disturbs carrier recombination and, as a consequence, lowers the optical transition probability [25–27]. To overcome this issue, Waltereit *et al* firstly proposed InGaN QWs on nonpolar ($1\bar{1}00$) plane free of internal electric fields, because the nonpolar planes are tilted by 90° from the conventional (0001) plane, as shown in Fig. 1.5, and involve the same numbers of anions and cations so that no electric field appears along the surface normal. On the other hand, semipolar planes, which are defined as planes inclined from the conventional polar (0001) plane by an angle (θ) between 0° and 90° , are also attractive. Several widely examined semipolar planes, $\{1\bar{1}01\}$, $\{11\bar{2}2\}$, and $\{20\bar{2}1\}$, are shown in Fig. 1.5. At the initial stage, a major hurdle in realizing nonpolar and semipolar light emitters was how to grow high-quality films because of the lack of suitable substrates.

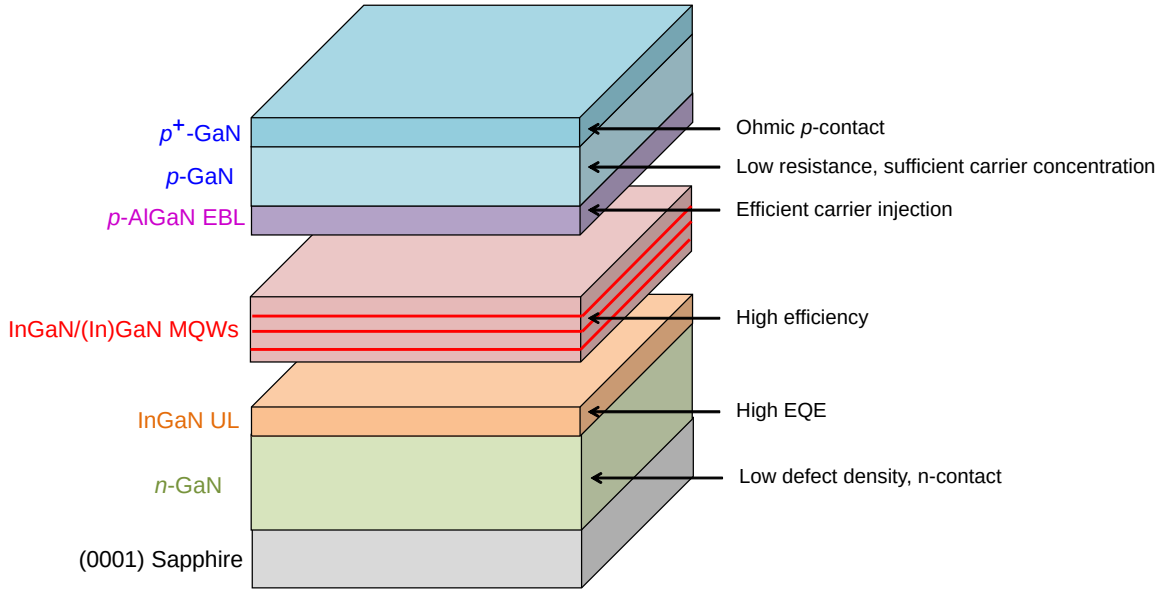


Figure 1.4: A typical InGaN-based LED structure and the requirements for each epi-layer.

Epitaxy on foreign substrates hindered the attractive semipolar and nonpolar physics, mainly due to the presence of the high dislocation densities. Excellent semipolar and nonpolar material qualities were obtained by the 3D faceted InGaN QWs via selective area growth. The 3D structure are composed of polar (0001), semipolar $\{11\bar{2}2\}$ and $\{1\bar{1}01\}$, and nonpolar $\{11\bar{2}0\}$ and $\{1\bar{1}00\}$ planes, thereby providing semipolar and/or nonpolar planes. InGaN QWs grown on semipolar and nonpolar microfacets successfully exhibited high emission efficiencies due to weakened internal electric fields [46, 47]. This finding motivated homoepitaxially-grown semipolar and nonpolar light emitters on tilted GaN substrates, which is sliced from (0001) GaN bulk crystals [28–42, 44, 45, 48, 49]. Figure 1.6 summarizes so far reported EQE for InGaN LEDs on semi- and non-polar planes. Although the EQE is still lower than that of (0001) InGaN LEDs, it is showing steady progress. One of the remarkable achievements of semipolar light emitters is higher output power operations of green LDs on $\{20\bar{2}1\}$ plane than those of conventional (0001) plane [50–55].

A continuing concern for nonpolar and semipolar light emitters is the high price of bulk GaN substrates compared with the commercially used sapphire substrates for polar (0001) LEDs. However, research and development are continuing to drive down the cost and improve the quality of these bulk GaN substrates. Han *et al.* have recently demonstrated semipolar GaN templates on 2-inch sapphire substrates with quality comparable to that of commercial (0001) GaN on sapphire substrates [56, 57]. This development

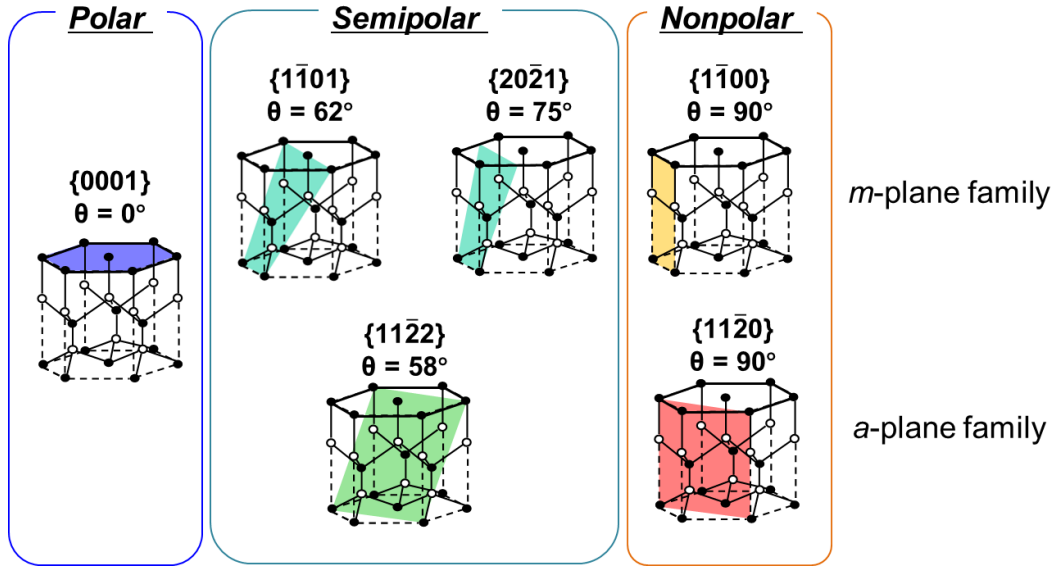


Figure 1.5: Schematics of (0001) polar plane, $(11\bar{2}0)$ and $(1\bar{1}00)$ nonpolar planes, and several widely investigated semipolar planes of III-nitride wurtzite crystal structures. The degrees indicate the off angles with respect to (0001) plane.

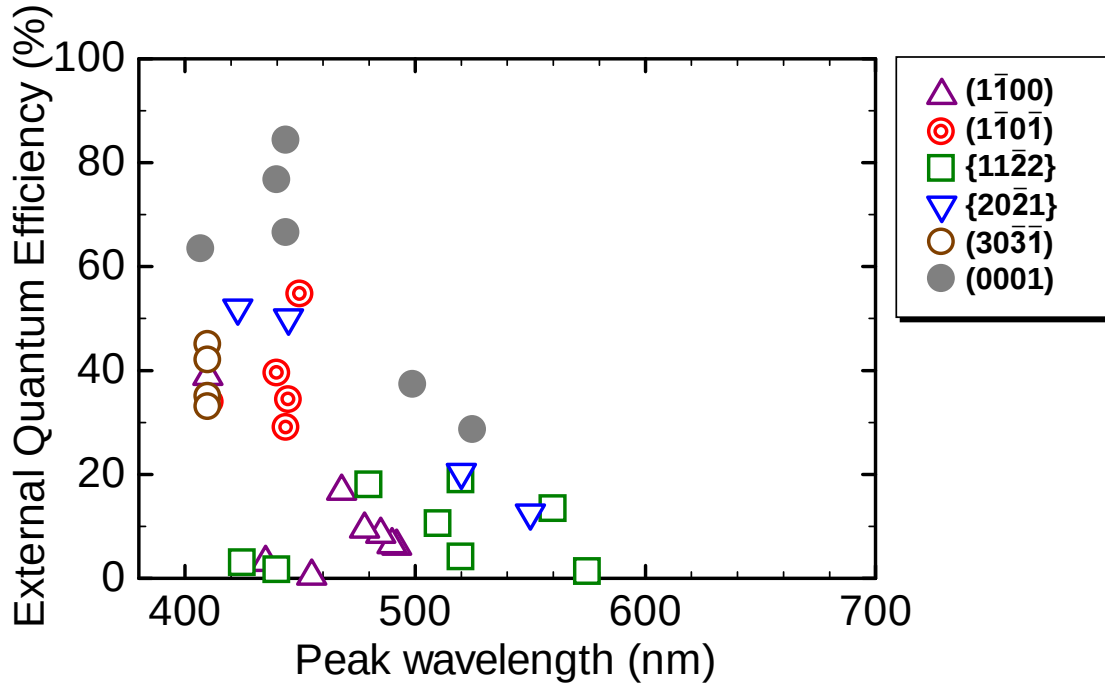


Figure 1.6: EQE as a function of emission wavelength for semi- and non-polar InGaN LEDs at a driving current of 20 mA under pulsed or direct current operations. $(1\bar{1}00)$ from Refs. [28–31], $(11\bar{0}\bar{1})$ from Refs. [32–36], $\{11\bar{2}\bar{2}\}$ from Refs. [37–41], $\{20\bar{2}\bar{1}\}$ from Refs. [42–44], $(30\bar{3}\bar{1})$ from Ref. [45]. As a reference, (0001) polar InGaN LEDs from Refs. [18, 20] (already shown in Fig. 1.3) are also indicated.

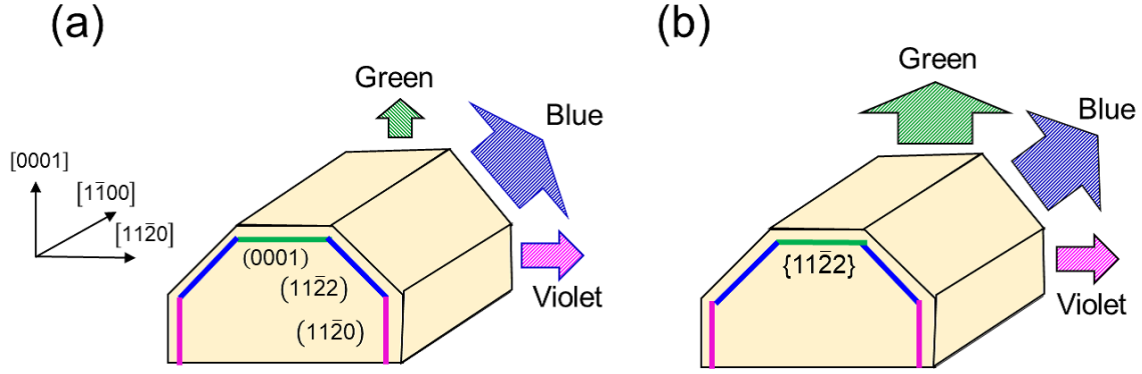


Figure 1.7: Schematics of 3D InGaN QWs on (a) (0001) and (b) $\{11\bar{2}2\}$

is expected to facilitate the future development and commercialization of semipolar LEDs.

Looking back at the 3D faceted InGaN LEDs, they also suffer from strain-induced polarization fields. So far reported structures have been usually fabricated on (0001) GaN templates, and the longest wavelength emission relies on the polar (0001) facet [4, 7, 10, 46, 58–60], as shown in Fig. 1.7(a). Nishizuka *et al.* experimentally observed internal quantum efficiency (IQE) of the $\{11\bar{2}2\}$ facet QWs was higher than that of the (0001) facet QWs, owing to the absence of internal electric field [46]. Similar trend was also reported by Ko *et al.* [61] and Funato *et al.* [62]. Therefore, the polarization issue in 3D faceted InGaN LEDs should be overcome by some novel ways toward highly efficient monolithic polychromatic emitters.

From the above background, efforts in this study are devoted to develop a novel 3D LED structure, which enables us to avoid the polarization issues. To explore such an advanced 3D structure, a semipolar $\{11\bar{2}2\}$ GaN substrate was employed for 3D structure growth. As previously reported by Ni *et al.* [63], 3D GaN structures on (11 $\bar{2}2$) GaN templates can be composed of $\{1\bar{1}01\}$ semipolar and $\{1\bar{1}00\}$ nonpolar facets without (0001) polar facet. This result indicates the possibility of realizing polychromatic emission from the 3D InGaN QWs without (0001) polar plane. Figure 1.7(b) illustrates such a promising 3D InGaN QWs on $\{11\bar{2}2\}$ plane. However, unlike them, homoepitaxy on $\{11\bar{2}2\}$ GaN substrates were selected. This is because, as described before, semipolar epitaxy on foreign substrates involves numerous dislocation densities and basal-plane stacking faults (BSFs). Ni *et al.* used *m*-plane sapphire substrates for (11 $\bar{2}2$) GaN, and the regrown 3D GaN structures still show poor material qualities originating from the GaN templates, which would prevent the overgrown semipolar QWs from exhibiting its full potential. Homoepitaxy can be expected to enable superior material quality of

regrown 3D GaN.

Following this chapter, chapter 2 describes sample preparation, MOVPE system, 3D GaN structure growth on $\{11\bar{2}2\}$ GaN substrates. Various stripe mask designs are formed on both $\pm(11\bar{2}2)$ GaN surface, and the overgrown 3D GaN structures are observed by cross-sectional scanning electron microscope (SEM). Then, the growth evolution is investigated, utilizing periodically inserted AlGaIn marker layers. We reveal that polar-plane-free faceted 3D structures can be fabricated on both $\pm(11\bar{2}2)$ GaN surface with a particular SiO_2 mask geometry. In chapter 3, InGaIn/GaN QWs are grown on the polar-plane-free 3D structures obtained in chapter 2, and the optical and structural properties are evaluated. The 3D faceted InGaIn QWs on $(\bar{1}\bar{1}2\bar{2})$ are demonstrated to exhibit radiative polychromatic emission properties. Chapter 4 is devoted to the investigation of pn-junction formation on each facet in the novel 3D structure on $(\bar{1}\bar{1}2\bar{2})$. In chapter 5, broadband electroluminescence from the polar-plane-free faceted 3D LEDs is demonstrated. The results of this study are summarized in chapter 6.

Finally, this is a little off-topic, but let us describe expected technological impacts of 3D facet InGaIn LEDs on micro-LED, which is currently a hot topic for display applications. Today, the conventional LED chip with the size of $\geq 200 \mu\text{m}^2$ has already been implemented in the display backlight module of liquid crystal displays (LCDs) or large size displays. On the other hand, micro-LED display integrates micro-LEDs with the size of less than $\sim 100 \mu\text{m}^2$ in arrays, as shown in Fig. 1.8. Current mainstream of flat panel displays (FPDs) are LCDs or organic LEDs (OLEDs). However, LCD requires an LED backlight to emit light through a matrix of liquid crystal, and it suffers from high power consumption, slow response time, and so on. On the other hand, OLED displays are self-emissive, but they have limited brightness and stability due to the inorganic nature. Compared with them, micro-LED displays possess potential advantages such as high contrast, fast response, low power consumption, and long lifetime.

At present, the micro-LED display technology still faces some challenges, like the transfer printing of micro-LED chips for mass production and the full-color method for display applications. In recent years, many companies have devoted considerable effort to develop transfer technologies by exploring many approaches [64]. On the other hand, full-color schemes are also a critical issue and are more involved with this study. The straightforward approach is to assemble discrete commercial LEDs with three different colors, and each pixel contains red, green, and blue LEDs. However, the advantages of high efficiency and long lifetime of conventional LED chips are inherited by miniatur-

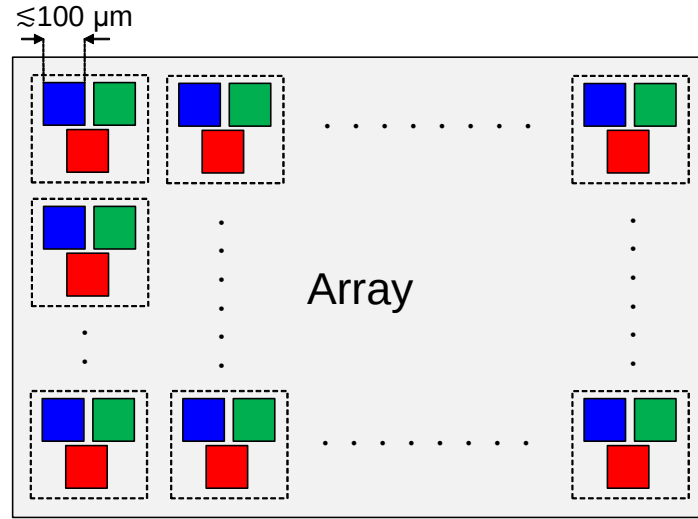


Figure 1.8: Schematic of micro-LED display.

ized one [65]. This degradation is mainly associated with the increased non-radiative surface recombination at the sidewall of the device, which is primarily introduced by a dry etching process. Additionally, the size gets smaller, the resolution enhances, but simultaneously elevating the fabrication complexity. A part of these complicated issues, however, might be solved by micro- or nano-structured 3D LEDs. First of all, chip transfer technologies are unnecessary for the 3D LEDs because the position-controlled selective area growth technique spontaneously arrays the polychromatic LEDs. At the same time, this implies that the etching process inducing the surface recombination can also be excluded from the fabrication procedure. One of the potential issues particular to the 3D LEDs would be electrode formation: due to the three-dimensionality and small dimensions around micron or submicron orders, it is challenging to form electrodes separately on each crystallographic facet.

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Chapter 2

3D Faceted GaN Structures on $\{11\bar{2}2\}$ GaN Substrates

2.1 Introduction

Polychromatic emission characteristics from three-dimensionally (3D) faceted InGaN quantum wells (QWs) strongly depend on the underlying 3D GaN structures. In this section, so far reported discussions on the origins of the polychromatic emission are introduced, and we define the favorable 3D GaN shapes.

Firstly, the origins of polychromatic emission properties of 3D faceted InGaN QWs are briefly explained. Figure 2.1(a) shows an example of cross-sectional scanning electron microscopy (SEM) image of 3D GaN on (0001) GaN template, and typical distribution of the QW structural parameters above the structure is designated. The InGaN

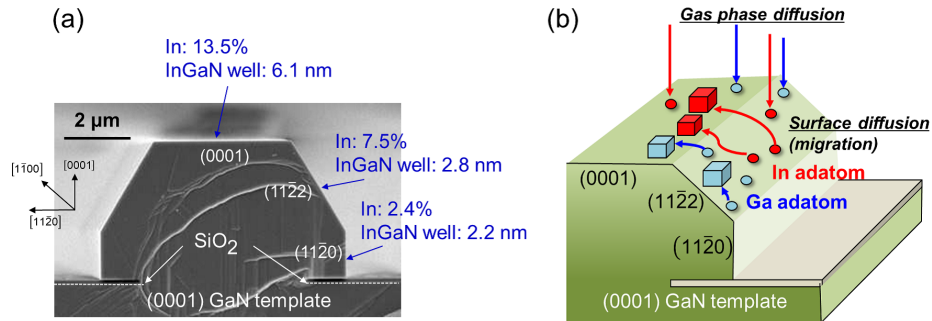


Figure 2.1: (a) Cross-sectional SEM image of 3D InGaN QWs on (0001) GaN template and the local QW structural properties. The structural parameters are cited from Ref. [1]. (b) Schematic illustration of growth dynamics of the InGaN QWs.

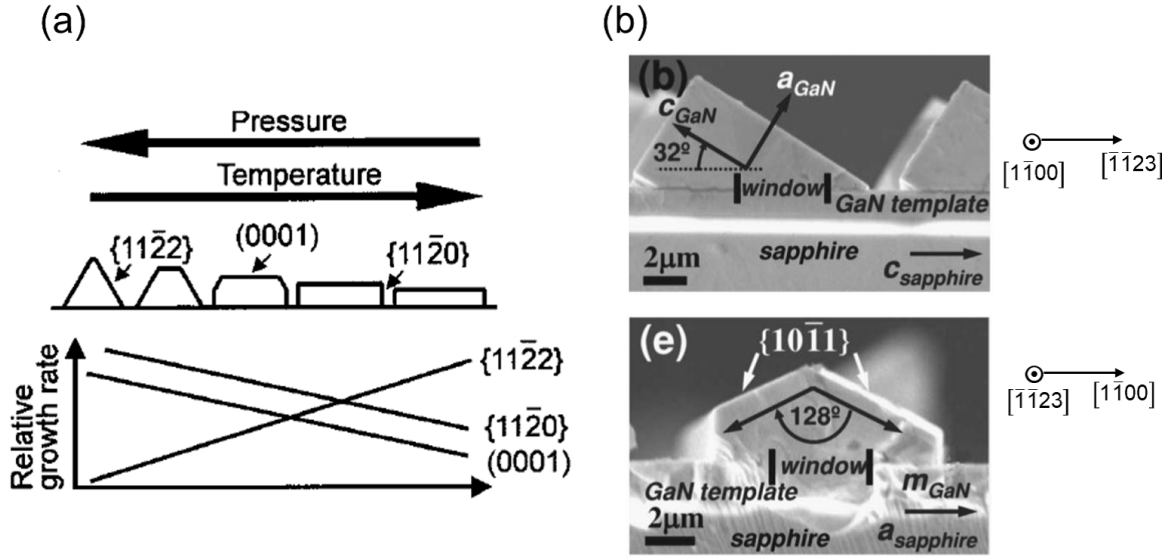


Figure 2.2: 3D GaN structures on (a) (0001) and (b) (11 $\bar{2}2$) cited from Refs. [6] and [8], respectively.

well widths and In compositions are cited representatively from Ref. [1]. Both the In compositions and well widths of the InGaN well vary in the order of the top (0001) > inclined (11 $\bar{2}2$) > side (11 $\bar{2}0$) facets. This variation results in the facet-dependent multi-wavelength emissions; the emission wavelength becomes longer from the side (11 $\bar{2}0$) to top (0001) facets. Ueda *et al.* discussed growth dynamics of the inter-facet variations of the InGaN QWs, and they found that surface adatoms migrate from {11 $\bar{2}2$ } to (0001) facet, and more In adatoms migrate than Ga adatoms do, which may cause the distribution of the local thickness and In composition in the 3D InGaN QWs [2]. More details will be discussed in chapter 3, compared with 3D InGaN QWs on {11 $\bar{2}2$ }. From these considerations, we can expect that trapezoidal 3D shapes with top and inclined facets are favorable for introducing polychromatic emission characteristics into the overgrown InGaN QWs.

Fabrication condition dependences of 3D GaN shapes on (0001) GaN templates have been extensively studied [3–7]. This is because selective area growth (SAG) and/or epitaxial lateral overgrowth (ELO) of 3D GaN structures based on metal-organic vapor phase epitaxy (MOVPE) can be useful to fabricate high-quality GaN epitaxial layers with low dislocation densities. Example 3D shape variation cited from Ref. [6] are shown in Fig. 2.2(a). The 3D GaN shapes change depending on the growth temperatures or growth pressures, and trapezoidal 3D shapes can be realized in particular growth conditions. On the other hand, little is known about regrowth of 3D GaN structures

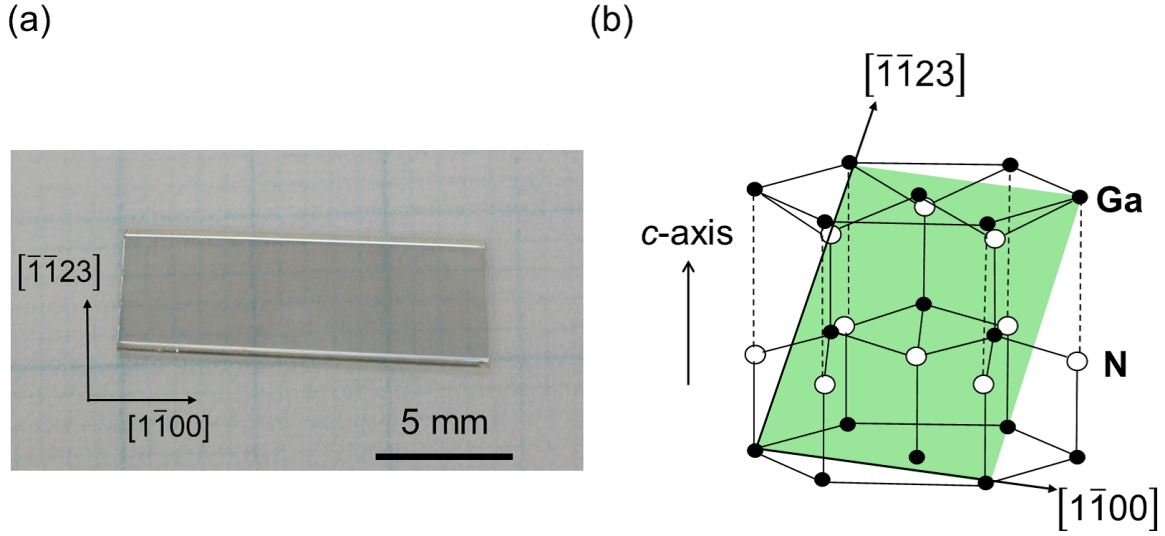


Figure 2.3: (a) An example photograph of a semipolar $\{11\bar{2}2\}$ GaN substrate used in this study. (b) Schematic of the $\{11\bar{2}2\}$ plane with respect to the GaN wurtzite lattice.

on semipolar planes. Within the limited papers, Ni *et al.* reported ELO-GaN on $(11\bar{2}2)$ GaN templates prepared by *m*-plane sapphire substrates [8]. The observed 3D structures are shown in Fig. 2.2(b), and the shapes close to triangles are disagree with our target 3D structure. Namely, fabrication conditions to obtain trapezoidal GaN structures on $\{11\bar{2}2\}$ plane remain uncertain.

In this study, we demonstrate that polar-plane-free faceted 3D GaN structures with trapezoidal shapes can be fabricated on $\{11\bar{2}2\}$ GaN substrates, by optimizing surface orientations and SiO_2 mask geometries. In addition, growth evolutions of the 3D GaN structures are discussed to get a deeper insight into the growth dynamics. The observed growth evolutions indicate that inter-facet surface adatom migration affects the growth developments of 3D shapes.

2.2 Sample Preparation

In this section, sample preparation procedures are briefly described. The process is commonly used throughout this study.

Figure 2.3 shows (a) an example photograph of a semipolar $\{11\bar{2}2\}$ GaN substrate used in this study and (b) $\{11\bar{2}2\}$ crystallographic plane with respect to the GaN wurtzite lattice. The substrates, which were collaboratively made by Furukawa Company, were sliced from a *c*-oriented GaN bulk crystal grown on sapphire (0001)

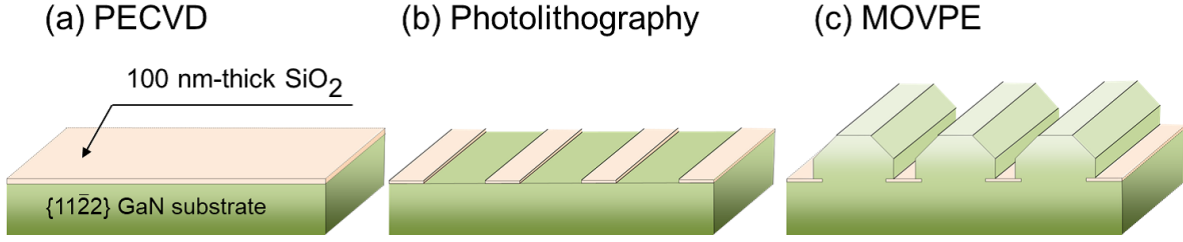


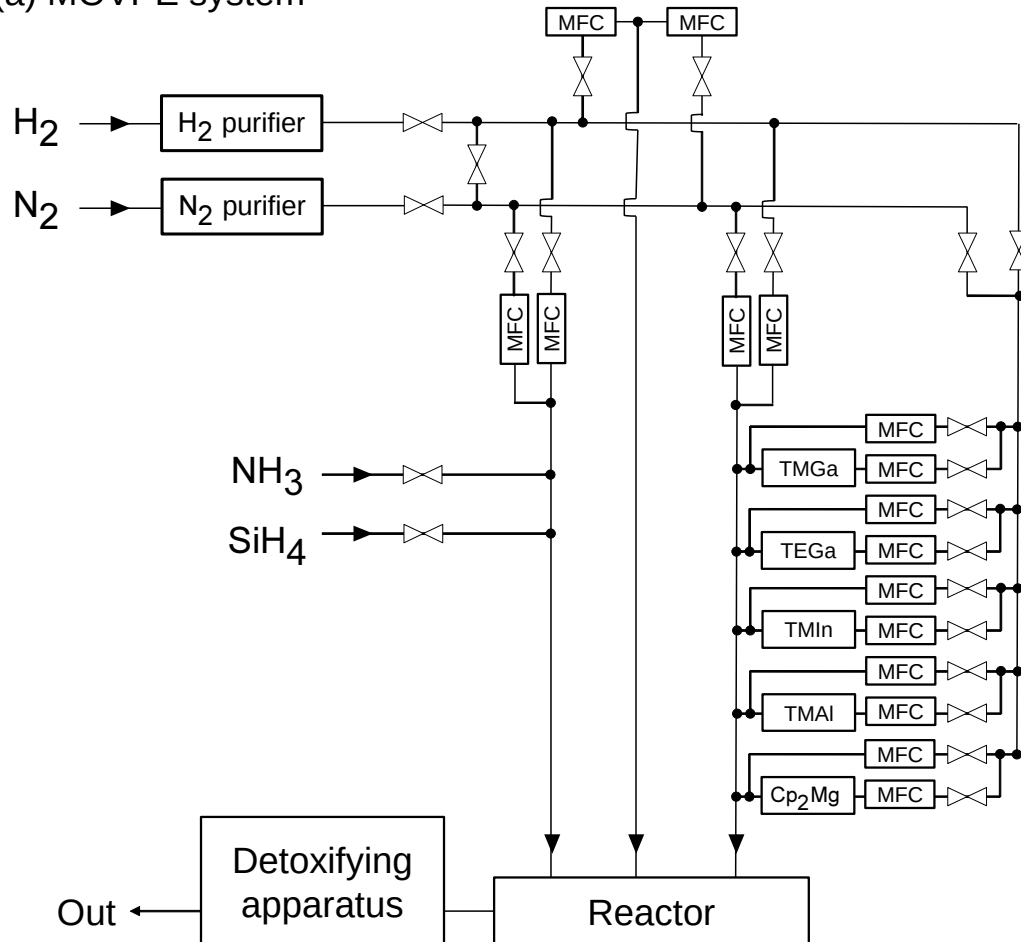
Figure 2.4: Schematic illustration of sample fabrication. (a) 100-nm-thick SiO₂ is deposited on a $\{11\bar{2}2\}$ GaN substrate by PECVD, then (b) striped SiO₂ mask is formed by UV photolithography and wet etching, and finally (c) 3D GaN structures are grown by MOVPE method.

by hydride vapor-phase epitaxy. Both the $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ surfaces are chemically and mechanically polished and epi-ready. The dislocation densities estimated by X-ray diffraction measurements are $\sim 10^6/\text{cm}^2$ for edge dislocation and $\sim 10^5/\text{cm}^2$ for screw dislocation. The substrates were intentionally doped with Si and possessed n-type conductivity with a typical carrier density of $\sim 10^{18}/\text{cm}^3$.

Figure 2.4 depicts fabrication procedures of 3D GaN structures. Striped SiO₂ mask was formed by plasma-enhanced chemical vapor deposition (PECVD) and UV photolithography. As-received $\{11\bar{2}2\}$ GaN substrates were firstly cleaned by decreasing in acetone, methanol, and purified H₂O. Then, 100-nm thick SiO₂ was deposited on the substrates by PECVD. Here, we note that the SiO₂ thickness was confirmed by spectroscopic ellipsometry. A standard photolithography process followed by a wet etch in buffered HF solution was used to define striped SiO₂ mask geometries. The samples were then pasted on 2-inch sapphire substrates with AlN paste. This is because, the sample holder of the MOVPE system used in this study was designed to a 2-inch wafer, as will be described later.

In order to explore the target 3D GaN structures, we focused on three parameters; (a) surface polarity, (b) stripe direction, and (c) mask geometry. For surface polarity, both the $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ surfaces were used, and then, two stripe directions of $[\bar{1}\bar{1}23]$ and $[1100]$ were prepared. The combinations of stripe window width w (μm) and mask width m (μm) were $(w, m) = (5, 5), (5, 15), (5, 45), (5, 95)$ and $(15, 5)$. The fill factor (FFs), which were defined as $w/(w+m)$, of these masks were 0.50, 0.25, 0.10, 0.05, and 0.75, respectively.

(a) MOVPE system



(b) Reactor

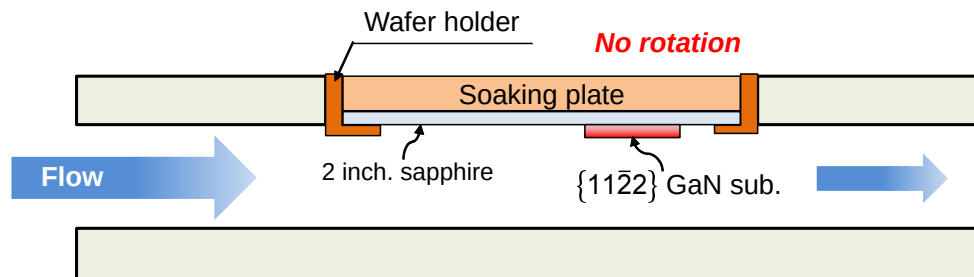


Figure 2.5: (a) Schematic diagram of the MOVPE system and (b) schematic model of the MOVPE reactor.

2.3 MOVPE Growth

The epitaxial growth was carried out using an MOVPE system. The MOVPE system is schematically illustrated in Fig. 2.5(a). Trimethylgallium (TMGa) and NH_3 were used as source precursors for GaN growth. H_2 was used as a carrier gas. The flow rates were precisely controlled by mass flow controllers. The cation sources (TMGa) and the anion sources (NH_3) were separately transferred from the bubblers to the reactor by H_2 passed through a Pd hydrogen purifier, and mixed at the very entrance of the reactor. The reactor was made of transparent quartz and was in the horizontal configuration.

Substrates were set on a SiC-coated wafer holder and a soaking plate. The substrate temperature was monitored using thermocouples. The difference between the measured and the real temperature was adjusted in advance using optimal growth condition of (0001) GaN on sapphire substrates. The measured temperatures which provide atomically flat GaN surfaces are attributed to 1050 °C, and the difference between the measured and the real temperature at room temperature is assumed to be zero. Figure 2.5(b) shows a schematic diagram of sample setting. The $\{11\bar{2}2\}$ GaN substrates used in this study were rectangular pieces, whereas the sample holder was designed to 2-inch wafers. Therefore, the $\{11\bar{2}2\}$ GaN substrates pasted on 2-inch sapphire substrates were set on the reactor.

Optimal growth conditions of homoepitaxial GaN films on $\{11\bar{2}2\}$ GaN substrates, which were revealed by Ueda *et al.* [9], were employed to the 3D GaN growth. Figure 3.2 shows (a) a structure design on planar surface and (b) a temperature profile used in this study. The substrates were initially annealed in NH_3 atmosphere, and homoepitaxial GaN was grown at 980 °C, which is lower than the optimal growth temperature (1050 °C) for conventional *c*-oriented GaN on sapphire. A clue to understanding why a lower temperature is suitable for growth on the $\{11\bar{2}2\}$ plane can be found in regrowth of 3D GaN structures on (0001) GaN where the $\{11\bar{2}2\}$ facets tend to appear at low temperature [5, 7] [see also Fig. 2.2(a)]. The reactor pressure was set to 300 Torr. The typical flow rates for TMGa and NH_3 were 69.3 $\mu\text{mol}/\text{min}$ and 2.1×10^2 mmol/min , respectively, yielding the V/III ratio of 3000. Under this growth condition, homoepitaxial GaN on both $\pm(11\bar{2}2)$ planar region typically yields a thickness of ~ 2.8 μm . Before the growth of 3D GaN structures, atomically flat surfaces of homoepitaxially-grown $\pm(11\bar{2}2)$ GaN were confirmed by atomic force microscopy (AFM).

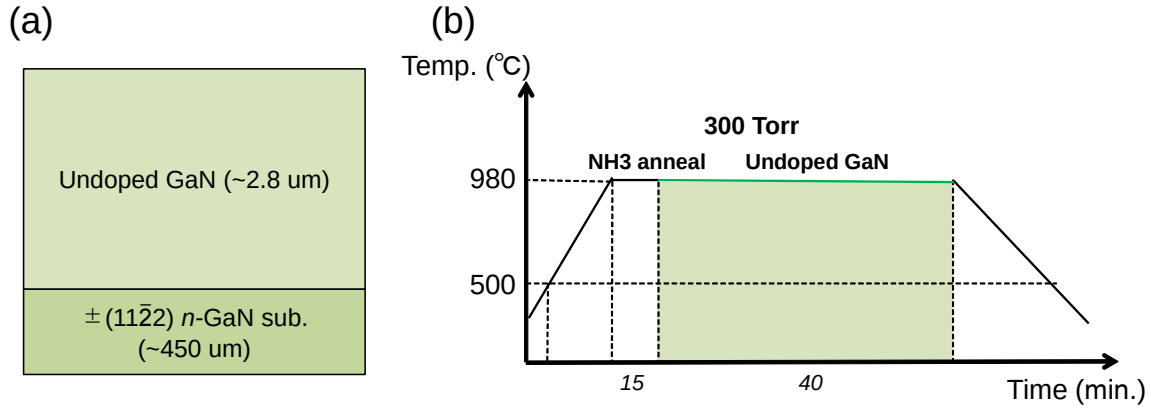


Figure 2.6: (a) A schematic structure in planar region and (b) a temperature profile of regrowth GaN.

2.4 3D GaN Structures on $\{11\bar{2}2\}$

Fabricated 3D GaN structures were observed by SEM using a JSM JEOL-6500F system operated at 5 kV. Figure 2.7 summarizes cross-sectional SEM images of 3D GaN structures overgrown on mask patterns with FFs of 0.50 and 0.75. The mask patterns with smaller FFs (0.05 – 0.25) provide similar structures to those with $FF = 0.50$, though the dimensions depend on FFs. Therefore, $FF = 0.50$ and 0.75 are representatively used in Fig. 2.7. With the $[\bar{1}\bar{1}23]$ stripes on the $(11\bar{2}2)$ surface, the 3D structures are triangle or trapezoidal shapes composed of $\{1\bar{1}01\}$ and $(11\bar{2}2)$. These planes have also been observed in selective area growth of GaN on the (0001) plane [3–7]. The inversed $\{1\bar{1}0\bar{1}\}$ and $(\bar{1}\bar{1}2\bar{2})$ planes and additional side $\{1\bar{1}00\}$ facets appear on the $(\bar{1}\bar{1}2\bar{2})$ surface with the same directional stripes. These planes have been observed in selective area growth on semipolar planes [10]. On the other hand, with the $[1\bar{1}00]$ stripes, the cross-sectional shapes are triangles, trapezoids, or inversed trapezoids composed of $(11\bar{2}2)$ and inclined $(000\bar{1})$ and $(\bar{1}\bar{1}22)$ planes. The $(000\bar{1})$ planes have also been found in selective area MOVPE of *a*- or *m*-plane GaN [11–15]. According to the above results, the exposed crystallographic orientations in Fig. 2.7 are considered to be often observed in selective area MOVPE of GaN.

Especially with the $[\bar{1}\bar{1}23]$ stripe and an FF of 0.75 on the $(11\bar{2}2)$ surface (surrounded by blue in Fig. 2.7), the 3D GaN structure is trapezoidal shape composed of the $(11\bar{2}2)$ and $\{1\bar{1}01\}$ semipolar planes. The opposite $(\bar{1}\bar{1}2\bar{2})$ plane with the same mask geometry (surrounded by red in Fig. 2.7) exhibits a similar structure composed of the $(\bar{1}\bar{1}2\bar{2})$ and $\{1\bar{1}0\bar{1}\}$ semipolar planes and $\{1\bar{1}00\}$ nonpolar planes. That is, these 3D structures

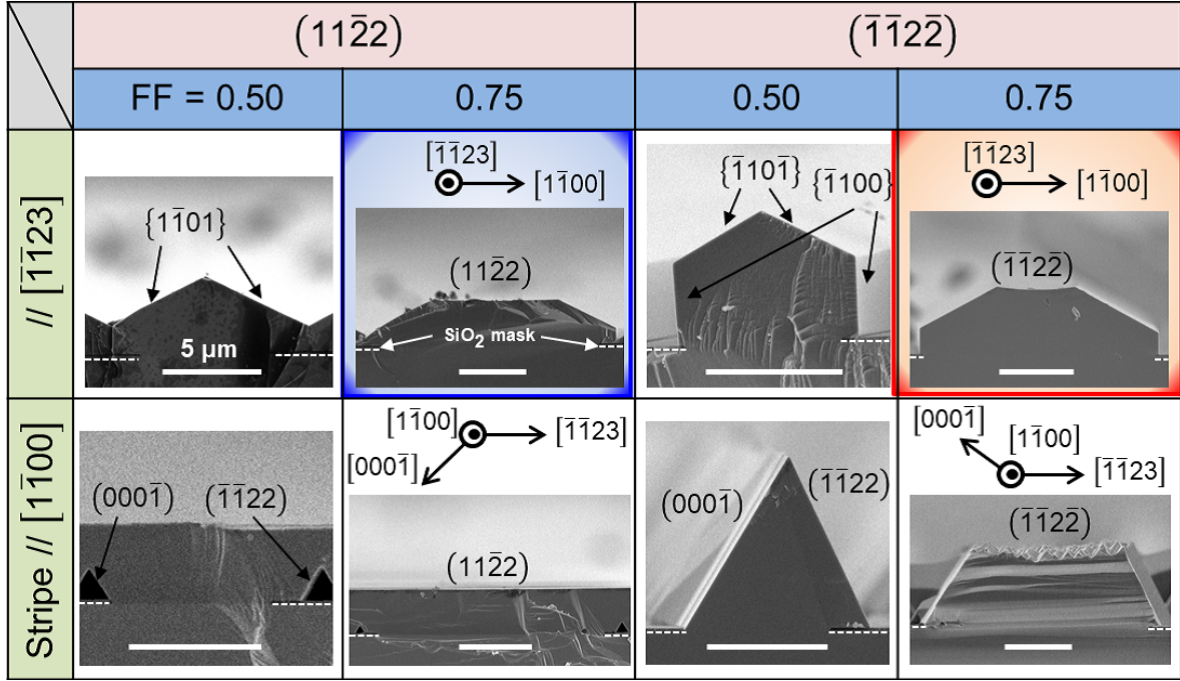


Figure 2.7: Cross-sectional SEM images of 3D-GaN structures. Scale bars represent 5 μm .

exclude the (0001) polar plane and meet the requirement of the target structures, which can be expected to exhibit polychromatic emissions. Hereafter, the discussions are focused on these two structures.

2.5 Growth Processes of 3D GaN Structures

In this section, growth processes of the polar-plane-free 3D GaN structures on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ are investigated. Although we have found the fabrication conditions to obtain these two structures, the growth dynamics behind them are unclear. In this situation, it is difficult to tailor the 3D shapes to meet some requirements. For example, a relative intensity ratio of longer and shorter wavelength emissions from 3D InGaN QWs depends on the originating facet area. Ueda *et al.* experimentally demonstrated that the emission spectra from the 3D QWs on (0001) can be tuned by controlling 3D GaN structures via altering SiO_2 mask geometries [16]. Therefore, getting an insight into the growth dynamics of the 3D GaN structures is indispensable for controlling emission characteristics.

Here, let us briefly summarize fundamental physics and current status of researches

related to growth shapes. Under equilibrium conditions, the growth shape can be determined by convexifying the plot of surface free energy, γ , versus surface normal, which is well known as $\gamma(\theta, \phi)$ -plot or simply γ -plot [17]. This is equivalent to minimizing the total free energy with respect to the total surface free energy under mass conservation, namely the Wulff construction. In this case, the growth shape should keep similar figures during the growth, though the dimension becomes larger. In practical, non-equilibrium conditions like MOVPE, the Wulff plot is replaced by the kinetic Wulff plot (v -plot) proposed by Du *et al.* [18], Sun *et al.* [11], and Jindal *et al.* [12] in GaN systems. This is because, surface processes such as adsorption, desorption, and surface diffusion also play an important role in determining the growth shape. Semi-empirical function, $v(\theta, \phi)$, where $v(\theta, \phi)$ is the growth velocity (v) of a plane with a surface normal defined by the polar (θ) and the azimuthal (ϕ) angles, substitutes the usual Wulff plot. The kinetic Wulff plot is constructed with information on the growth velocity of different orientations, obtained experimentally. In this case, growth velocity of each facet is assumed to be independent of time. However, the above discussions are focused on the final growth shapes after the epitaxy, and on-going shapes are hidden. This situation prevents us from understanding which factors determine the growth velocity of each facet. Therefore, this study attempts to visualize the temporal developments of 3D GaN structures and elucidate the background growth mechanism.

2.5.1 Growth evolutions visualized by AlGaIn markers

Accessing 3D shape evolutions during the growth gives us much information about the growth mechanism. Figure 2.8 shows example cross-sectional SEM images of marker-inserted 3D structures [19,20]. In Fig. 2.8(a), Shioda *et al.* studied growth rate profiles adjacent to selective area masks of InP with InGaAs markers, and found time-proportional growth rate increments of InP growth at the mask edge [19]. They also discussed the temporal development of the growth rates based on vapor phase diffusion and surface diffusion combined model. It must be noted that such a detailed investigation cannot be conducted by just observing the final growth shapes. In GaN systems, AlGaIn marker layers would be helpful to perform a similar analysis. This is because, the AlGaIn marker layers in GaN are well visible as in SEM images. Habel *et al.* experimentally demonstrated that AlGaIn layers provide direct access to the propagating growth facets in different crystal directions and under different growth conditions [20]. By use of this technique, they proposed multi-step facet assisted epitaxial lateral overgrowth (FACELO) toward low dislocation density GaN. However, their study was limited to

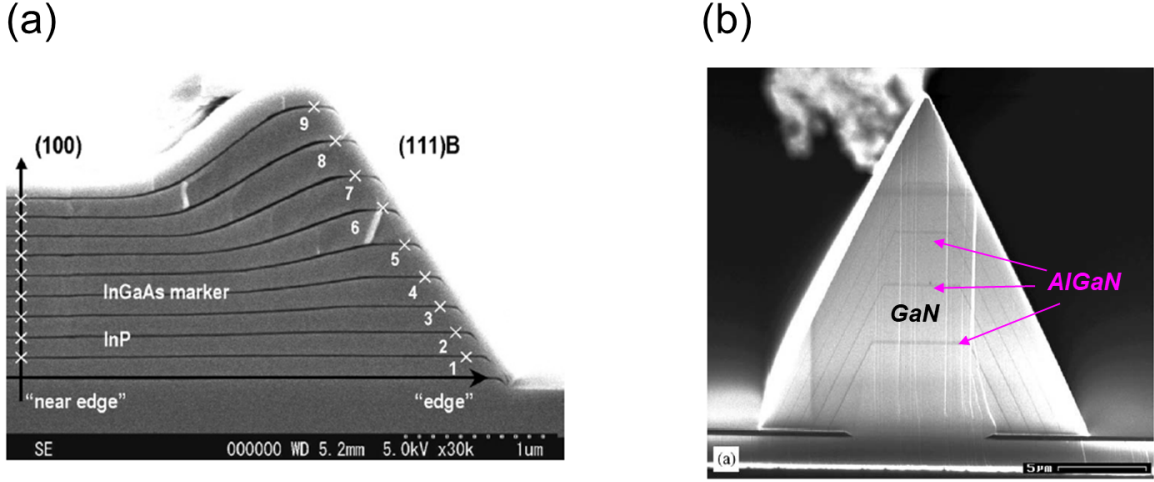


Figure 2.8: Cross-sectional SEM images of (a) InP multi-layer structure with InGaAs marker and (b) AlGaIn marker layers inserted in 3D GaN structure reproduced from Refs. [19] and [20], respectively.

the observations, and further analyses using AlGaIn markers have yet to be performed. Therefore, in this study, temporal developments of 3D GaN on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ are accessed by AlGaIn markers, and the temporal growth rate variations are quantitatively discussed in terms of inter-facet adatom migration.

Samples were grown on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ GaN substrates with essentially the same procedures as described in section 2.3. Figure 2.9(a) and 2.9(b) show a schematic of epilayer design on $\pm(11\bar{2}2)$ planar surfaces and a temperature profile of 3D GaN structures with AlGaIn markers, respectively. In addition to TMGa and NH_3 , trimethylaluminum (TMAI) was periodically added to the reactor to form a periodic AlGaIn layer. The designed Al composition and thickness of the inserted AlGaIn layers on a planar surface were set to 10% and ~ 30 nm, respectively. The growth conditions of undoped GaN layers were the same as Fig. 3.2. A cross-sectional SEM image of the fabricated planar GaN with AlGaIn markers on $(\bar{1}\bar{1}2\bar{2})$ plane is shown in Fig. 2.9(c). The AlGaIn layers could be detected as darker SEM contrasts, as previously reported by Habel *et al.* [20]. Here, the acceleration voltage and the electron beam current were increased to enhance the SEM contrast between GaN and AlGaIn.

Figures 2.10(a) and 2.10(b) show cross-sectional SEM images of fabricated 3D GaN structures with AlGaIn markers on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$, respectively. Each AlGaIn marker layer directly indicates the position and shape of the growing facet for the respective point in time. From the observation, it is found that the cross-sectional shape of 3D GaN on $(\bar{1}\bar{1}2\bar{2})$ drastically changes during the growth [Fig. 2.10(b)]. As the growth

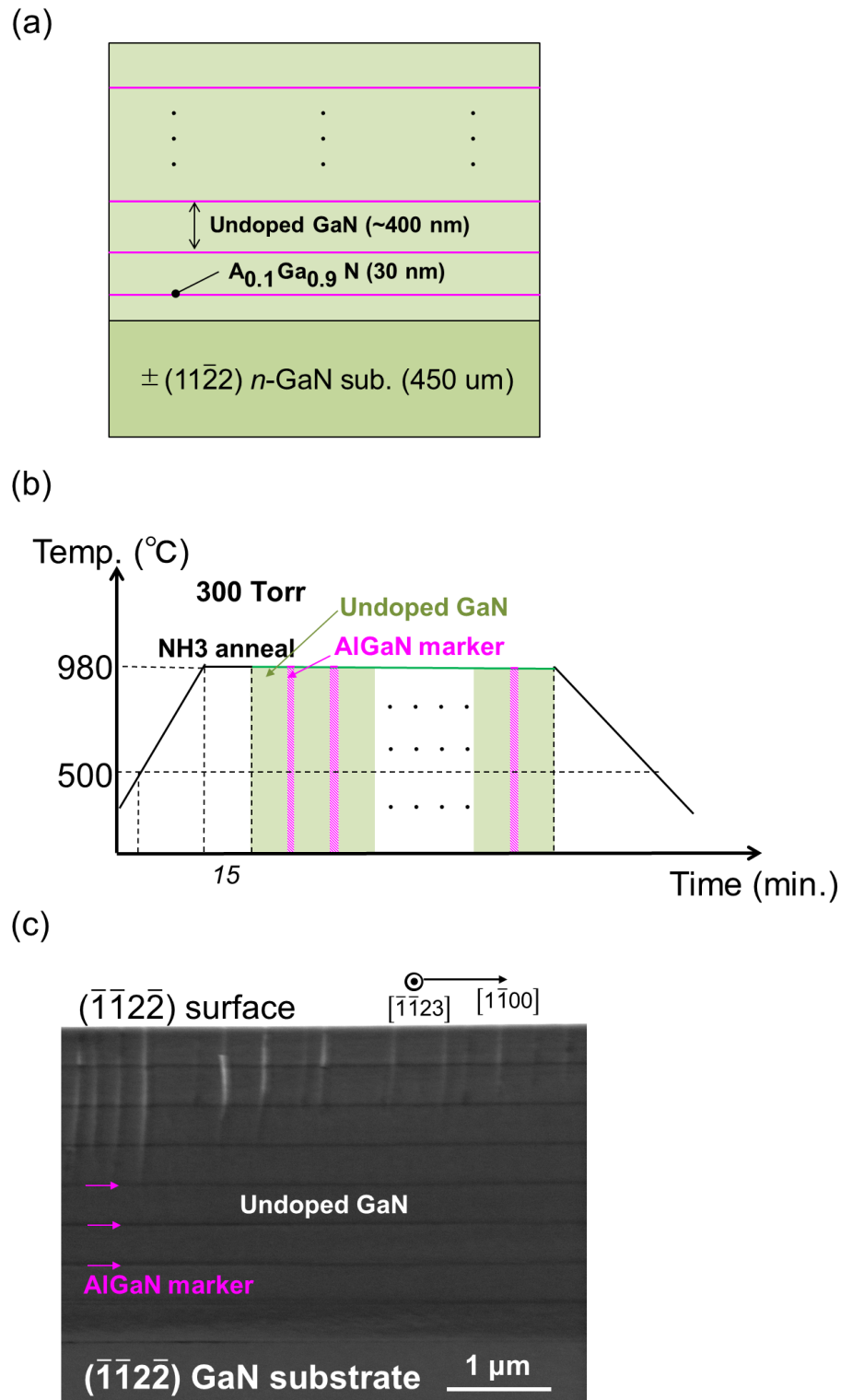


Figure 2.9: Growth condition of 3D GaN structures with AlGaIn markers. (a) A schematic structure design on planar surface and (b) a temperature profile. (c) An example cross-sectional SEM image of fabricated planar $(\bar{1}\bar{1}2\bar{2})$ GaN with AlGaIn markers.

proceeds, the top $(\bar{1}\bar{1}2\bar{2})$ facet becomes narrower, whereas the inclined $(\bar{1}10\bar{1})$ and side $(\bar{1}100)$ facets are widened. On the other hand, for the 3D GaN on $(11\bar{2}2)$ [Fig. 2.10(a)], the inclined $(1\bar{1}01)$ facet is widened, while the top $(11\bar{2}2)$ facet shrinks slowly compared with the top $(\bar{1}\bar{1}2\bar{2})$ facet of the 3D GaN on $(\bar{1}\bar{1}2\bar{2})$. Because these 3D structures have similar trapezoidal cross sections, the geometrically determined diffusion process in the vapor phase over them would be similar. Therefore, the different facet developments indicate distinct surface processes between them.

To quantitatively discuss the above-mentioned growth behaviors, the facet width and growth rate on each facet were measured at each growth step. The growth step numbers are indicated in Fig. 2.10(a) and Fig. 2.10(b). Figure 2.11 shows definitions of the facet width and growth rate on each facet, and the measured facet widths are summarized in Figure 2.10(c) and 2.10(d) as a function of the growth step. Here, total facet widths indicated as “Total” correspond to the entire facet widths of the 3D GaN: $\text{Total} = L_{\text{top}} + 2 \cdot L_{\text{inclined}} + 2 \cdot L_{\text{side}}$. “Pattern period” corresponds to the period of the mask pattern, which is $15 \mu\text{m}$ window width + $5 \mu\text{m}$ mask width = $20 \mu\text{m}$ for both 3D GaN. As expected from the observations, the top $(\bar{1}\bar{1}2\bar{2})$ facet width of the 3D GaN on $(\bar{1}\bar{1}2\bar{2})$ [Fig. 2.10(d)] decrease sharply compared to the top $(11\bar{2}2)$ facet width of the 3D GaN on $(11\bar{2}2)$. On the other hand, the inclined $(1\bar{1}01)$ and $(\bar{1}10\bar{1})$ facets of both the 3D GaN on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ show similar facet width increments during the growth. To further discuss the growth dynamics, growth rate evolutions at each growth step are compared.

Figures 2.10(e) and 2.10(f) summarize the growth rate developments as a function of the growth step. As a reference, the growth rates of maskless planar region are also indicated. The distinct growth rate evolutions are revealed from Figs. 2.10(e) and 2.10(f): as the growth proceeds, the growth rates of the top and inclined facets of the 3D GaN on $(11\bar{2}2)$ monotonically decrease, while those of the 3D GaN on $(\bar{1}\bar{1}2\bar{2})$ monotonically increase. In addition, viewed apart, Fig. 2.10(e) presents that the growth rates of the top $(11\bar{2}2)$ facet are near that of the planar $(11\bar{2}2)$ GaN, and the growth rates of the inclined $(1\bar{1}01)$ facet are slightly slower than those of the top $(11\bar{2}2)$ facet. On the other hand, Fig. 2.10(f) reveals that the growth rate of the top $(\bar{1}\bar{1}2\bar{2})$ facet is much larger than that of planar one, and the $(\bar{1}\bar{1}2\bar{2})$ growth rates are much faster than the $(\bar{1}10\bar{1})$ growth rates. These results suggest distinct growth dynamics behind the shape evolutions between the 3D GaN on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$. Here, we note that kinetic Wulff construction must be carefully used especially for growth shapes on $(11\bar{2}2)$, because the relative facet growth velocity is very time-dependent.

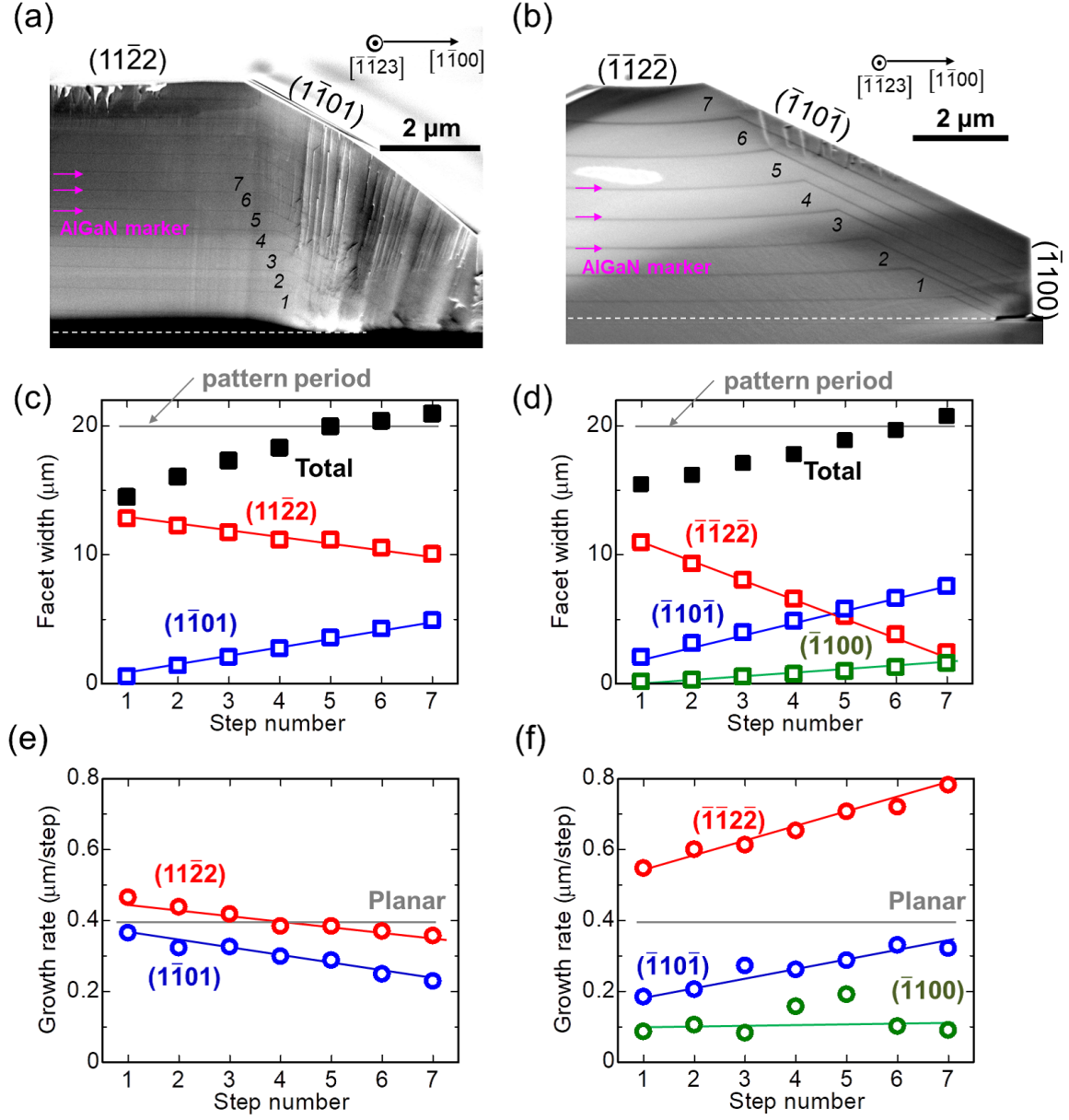


Figure 2.10: Cross-sectional SEM images of 3D-GaN structures with AlGaIn marker layers on (a) (11 $\bar{2}$ 2) and (b) ($\bar{1}$ 1 $\bar{2}$ 2), and (c, d) each facet width and (e, f) growth rate developments as a function of the growth step indicated in (a) and (b).

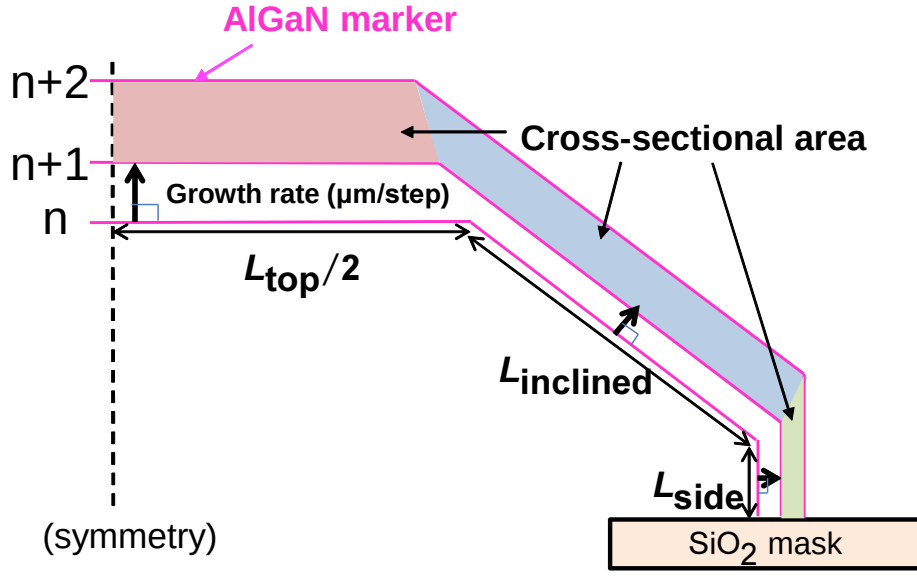


Figure 2.11: Definitions of facet width, growth rate, and cross-sectional area on each facet.

factors affecting facet growth rates

Generally, a growth rate can be approximated by $v \propto k \cdot J$, where the k and J are the incorporation efficiency and the sum of fluxes of atoms from the gas phase and adatoms migrating on the surface, respectively. Thus, a larger k and/or J provides a larger growth rate. Here, incorporation efficiency (k) of each crystallographic plane should be independent of growth duration. Therefore, the observed growth rate developments are attributed to the mass transport (J). Now, the total amount of precursors transported to the 3D GaN growth front from the gas phase and mask region could be assumed to be constant during the growth. In addition, it is worth noting that the sum of the cross-sectional areas of the top, inclined, and side facets represented in Fig. 2.11 was calculated to be nearly time-independent for both the 3D GaN on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$, which indicates that the amount of precursors incorporated to the 3D GaN is constant through all the growth steps. Therefore, the observed growth rate developments can be attributed to the inter-facet surface diffusion of adatoms between the top and inclined facets.

For the 3D GaN on $(11\bar{2}2)$ [Fig. 2.10(e)], the growth rates of the top $(11\bar{2}2)$ facet, $v_{(11\bar{2}2)}^{\text{facet}}$, at initial growth steps are slightly faster than those of the planar $(11\bar{2}2)$ GaN, $v_{(11\bar{2}2)}^{\text{planar}}$, indicating that the surface adatoms diffuse from the inclined into the top facets, in addition to the vapor phase diffusion into the top facet. However, $v_{(11\bar{2}2)}^{\text{facet}}$ decreases

and becomes slower than $v_{(11\bar{2}2)}^{\text{planar}}$ during the growth. One of the plausible reasons for this variation is increasing total facet width indicated in Fig. 2.10(c). As mentioned above, the total amount of precursors transported to the 3D GaN growth front from the gas phase and mask region is considered to be constant during the growth in this case. Thus, increasing growth front, *i.e.*, the total facet width, with constant total amount of precursors decrease the effective fluxes (J) on the growth front, eventually lowers the growth rate. Meanwhile, for the 3D GaN on $(\bar{1}\bar{1}2\bar{2})$ [Fig. 2.10(f)], $v_{(\bar{1}\bar{1}2\bar{2})}^{\text{facet}}$ is much faster than $v_{(\bar{1}\bar{1}2\bar{2})}^{\text{planar}}$, suggesting drastic inter-facet adatom diffusion from the inclined to top facets, which is consistent with much slower $v_{(\bar{1}10\bar{1})}^{\text{facet}}$. This is because, the adatom diffusion from the inclined to the top facet increase the adatom density on the top facet, simultaneously leading to the less adatom density on the inclined facet. Additionally, both the $v_{(\bar{1}\bar{1}2\bar{2})}^{\text{facet}}$ and $v_{(\bar{1}10\bar{1})}^{\text{facet}}$ monotonically increase in spite of the increasing growth front shown in Fig.2.10(d), in contrast to the 3D GaN on $(11\bar{2}2)$. This behavior cannot be well explained in this stage, and imply further considerations necessary. Therefore, in the following sections, numerical calculations on inter-facet adatom diffusion are performed, and the temporal variations of adatom diffusion are discussed.

2.5.2 A model calculation of mass transport

In order to understand the origins of the growth rate evolutions of the 3D GaN on $\pm(11\bar{2}2)$, the inter-facet surface diffusion were estimated from the experimental data (Fig. 2.10) by a simple model based on the one-dimensional inter-facet surface migration. Figure 2.12 schematically shows this model, where the parameters on the top and inclined planes are represented as subscripts, while those on the planar region and facets are designed by superscripts. In this model, the flux of adatoms from the inclined to top facet (J_m) is assumed, and the top and inclined facet growth rates depend on both gas phase diffusion and the inter-facet surface diffusion. On the other hand, the growth rate on planar region depends on only gas phase diffusion. Noting that J_m (as well as J) is defined as an area density, the flux into the top facet can be expressed as $J + J_m \frac{L_{\text{inclined}}}{L_{\text{top}}/2}$. From the relationship $v \propto k \cdot J$, we obtain

$$\frac{J_{\text{top}}^{\text{facet}}}{J_{\text{top}}^{\text{planar}}} = \frac{k_{\text{top}} \left(J + J_m \frac{L_{\text{inclined}}}{L_{\text{top}}/2} \right)}{k_{\text{top}} J} = 1 + \frac{L_{\text{inclined}}}{L_{\text{top}}/2} \cdot \frac{J_m}{J}. \quad (2.1)$$

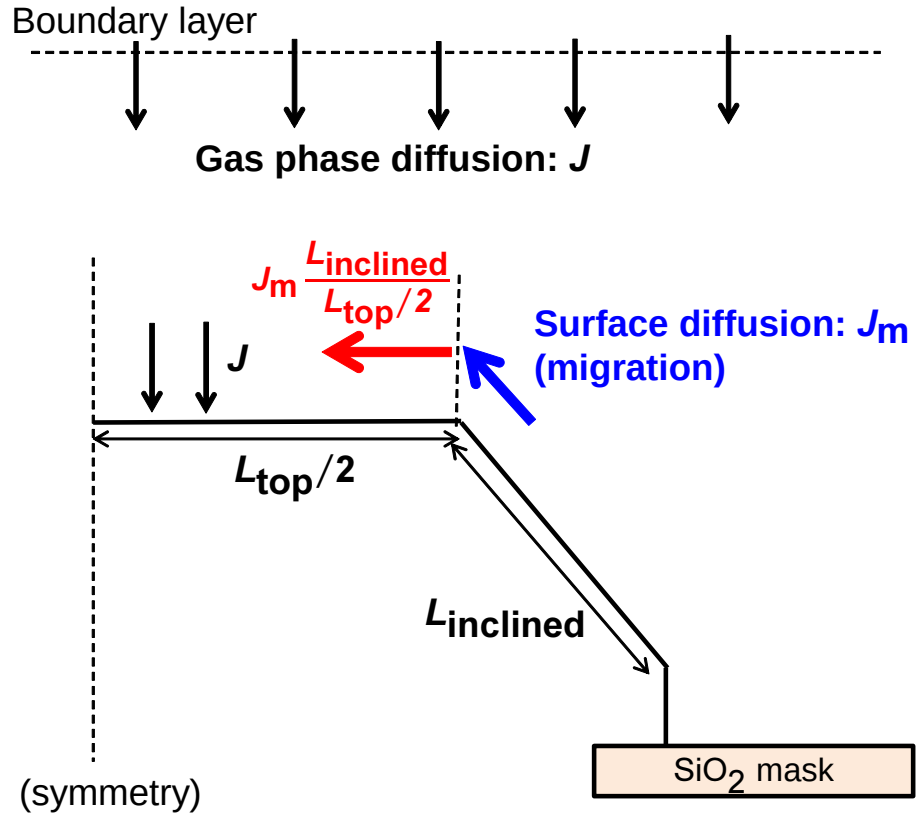
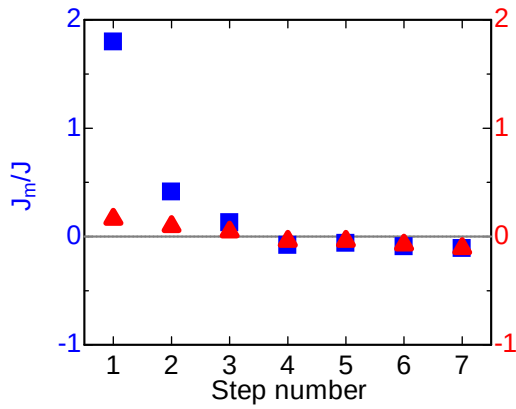
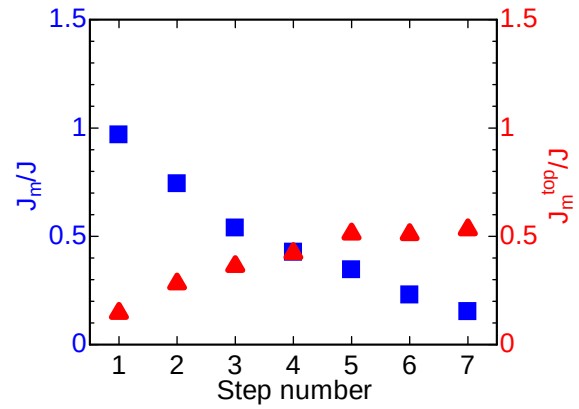


Figure 2.12: A model to simulate the growth dynamics of 3D GaN structures.

(a) 3D GaN on $(11\bar{2}2)$ (b) 3D GaN on $(\bar{1}\bar{1}2\bar{2})$ Figure 2.13: Normalized surface diffusion fluxes going out of the inclined facet (J_m/J) and coming into the top facet (J_m^{top}/J). 3D GaN on (a) $(11\bar{2}2)$ and (b) $(\bar{1}\bar{1}2\bar{2})$.

Solving this equation with Figs. 2.10(c)~2.10(f), the normalized flux of inter-facet surface diffusion J_m/J at each growth step was calculated.

Figure 2.13 shows the results, where $J_m^{\text{top}} = J_m \frac{L_{\text{inclined}}}{L_{\text{top}}/2}$ indicates the flux coming into the top facet. For the 3D GaN on $(11\bar{2}2)$ [Fig. 2.13(a)], as the growth proceeds, the J_m/J and J_m^{top}/J vary from the positive to negative values with decreasing the absolute values. Based on the definition, the positive J_m/J (negative) values suggest that the surface adatoms migrate from the inclined (top) to the top (inclined) facets, which is consistent with the growth rate development shown in Fig. 2.10(e). On the other hand, the 3D GaN on $(\bar{1}\bar{1}2\bar{2})$ exhibits the positive J_m/J and J_m^{top}/J values during the growth, corresponding to the much larger $v_{(\bar{1}\bar{1}2\bar{2})}^{\text{facet}}$ than $v_{(\bar{1}\bar{1}2\bar{2})}^{\text{planar}}$. Additionally, J_m^{top}/J increases in spite of decreased J_m/J . This is due to the term $\frac{L_{\text{inclined}}}{L_{\text{top}}/2}$. The top facet width L_{top} becomes narrower in contrast to the widened inclined facet width L_{inclined} , and thus the consequent $\frac{L_{\text{inclined}}}{L_{\text{top}}/2}$ drastically increases, which overcomes the decrease of J_m/J . These results well agree with the observation of growth rate increments of both the top and inclined facets; decreased J_m/J means more flux on the inclined facet, and increased $J_m^{\text{top}} = J_m$ obviously means increased flux on the top facet. From this discussion, the temporal growth rate variation is well accounted for by the temporal variation of inter-facet surface diffusion which is affected by the shape evolution of the 3D GaN structures.

2.5.3 Adatom kinetics on different surface orientations

Now, a question arises why the distinct inter-facet surface diffusion was indicated between the 3D GaN on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$. It should be noted that angles between the top and inclined facets for both structures are the same, so the attention should be directed to adatom diffusion on each crystallographic plane. One plausible explanation is that adatoms on $(\bar{1}\bar{1}2\bar{2})$ and $(\bar{1}10\bar{1})$ surfaces of the 3D GaN on $(\bar{1}\bar{1}2\bar{2})$ migrate more actively than those on $(11\bar{2}2)$ and $(1\bar{1}01)$ surfaces of the 3D GaN on $(11\bar{2}2)$. In the case of GaN MOVPE, surface bond configurations play a crucial role in the adatom diffusion dynamics on that plane. Zywietz *et al.* calculated the potential energy surface (PES) for Ga and N atoms on the $\pm(0001)$ surface, giving immediate insight into stable sites, migration paths, and diffusion barriers [21]. As a result, they pointed out a very different diffusivity for adatoms on (0001) and $(000\bar{1})$ surfaces: Ga adatom diffusion on N-terminated $(000\bar{1})$ GaN surface is more limited than that on N-terminated (0001) GaN surface. Therefore, $\pm(11\bar{2}2)$ and $\pm(1\bar{1}01)$ may also exhibit distinct Ga adatom diffusivity. However, at present, little is known about Ga adatom diffusivities on them

in terms of the PES calculation. To our knowledge, Ga adatom kinetics on only $(11\bar{2}2)$ GaN surface is discussed by Akiyama *et al.* [22]: the energy barrier for Ga migration on $(11\bar{2}2)$ surface (1.2 eV) is much higher than that on (0001) surface (~ 0.4 eV). In addition to the very limited report, the PES requires *ab initio*-based total-energy calculations, and thus it is difficult to expand the results intuitively to other surface orientations. Further experimental and theoretical studies are needed, but in the following discussion, we attempt to explain the adatom diffusivity based on an “ideal” GaN structure and the terminations of NH_i ($i = 1, 2, 3$) molecules covering Ga atoms.

The different atomic arrangements of $\pm(0001)$, $\pm(11\bar{2}2)$, and $\pm(1\bar{1}01)$ surfaces are presented in Figure 2.14. Let us discuss adatom diffusivity using these ideal structures. First of all, bare Ga adatoms on a surface are suggested to be unstable in terms of the surface phase diagram based on chemical potential calculations, and are likely to be stabilized by NH_i molecules termination particularly under N-rich MOVPE growth condition [23–27], where the source of molecular H is the H_2 carrier gas, NH_3 and TMGa. This indicates that the stability of the NH_i layer affects the Ga adatom diffusivity: stable NH_i termination suppresses the Ga adatom migration. Moreover, the NH ($i = 1$) termination is the most stable among NH_i layers, because the N atom forms three bonds to the underlying Ga atoms allowing better sticking. In other words, surface orientations covered by NH layers should exhibit limited Ga adatom diffusivities. Such a situation seems to be coincidence with $(000\bar{1})$ surface: as you can see in Fig. 2.14(a), the N atom form three bonds to the upside Ga atoms, and the NH termination over wide range of conditions is also theoretically predicted by Northrup *et al.* [24] and Akiyama *et al.* [27]. Experimentally, $(000\bar{1})$ GaN tends to form a rough surface including many hillocks under conditions similar to the (0001) GaN. Vicinal substrates [28] or surfactant effect [29, 30] have been applied to prepare the flat surfaces, indicating the less Ga adatom migration than that on (0001) surface.

The different Ga adatom diffusivity of $\pm(11\bar{2}2)$ and $\pm(1\bar{1}01)$ could be also explained by the analogy with (0001) vs. $(000\bar{1})$ GaN surfaces. For $\pm(11\bar{2}2)$ [Fig. 2.14(b)], the N atom on $(11\bar{2}2)$ surface forms two bonds to the underlying Ga atom, whereas that on $(\bar{1}\bar{1}2\bar{2})$ does one bond. Thus, the more stable NH_i termination is expected on $(11\bar{2}2)$ surface. Theoretically, Akiyama *et al.* predicted the $\text{NH} + \text{NH}_2$ termination of $(11\bar{2}2)$ surface over wide range of conditions, and pointed the surface polarity similar to $(000\bar{1})$ GaN surface [26, 27]. For $\pm(1\bar{1}01)$ [Fig. 2.14(c)], the N atom on $(1\bar{1}01)$ surface forms two or three bonds to the underlying Ga atom depending on the sites, whereas that on $(\bar{1}\bar{1}0\bar{1})$ does one or two bonds, suggesting more stable NH_i termination on the $(1\bar{1}01)$ surface. Akiyama *et al.* calculated such a stable termination of $(1\bar{1}01)$ over wide range

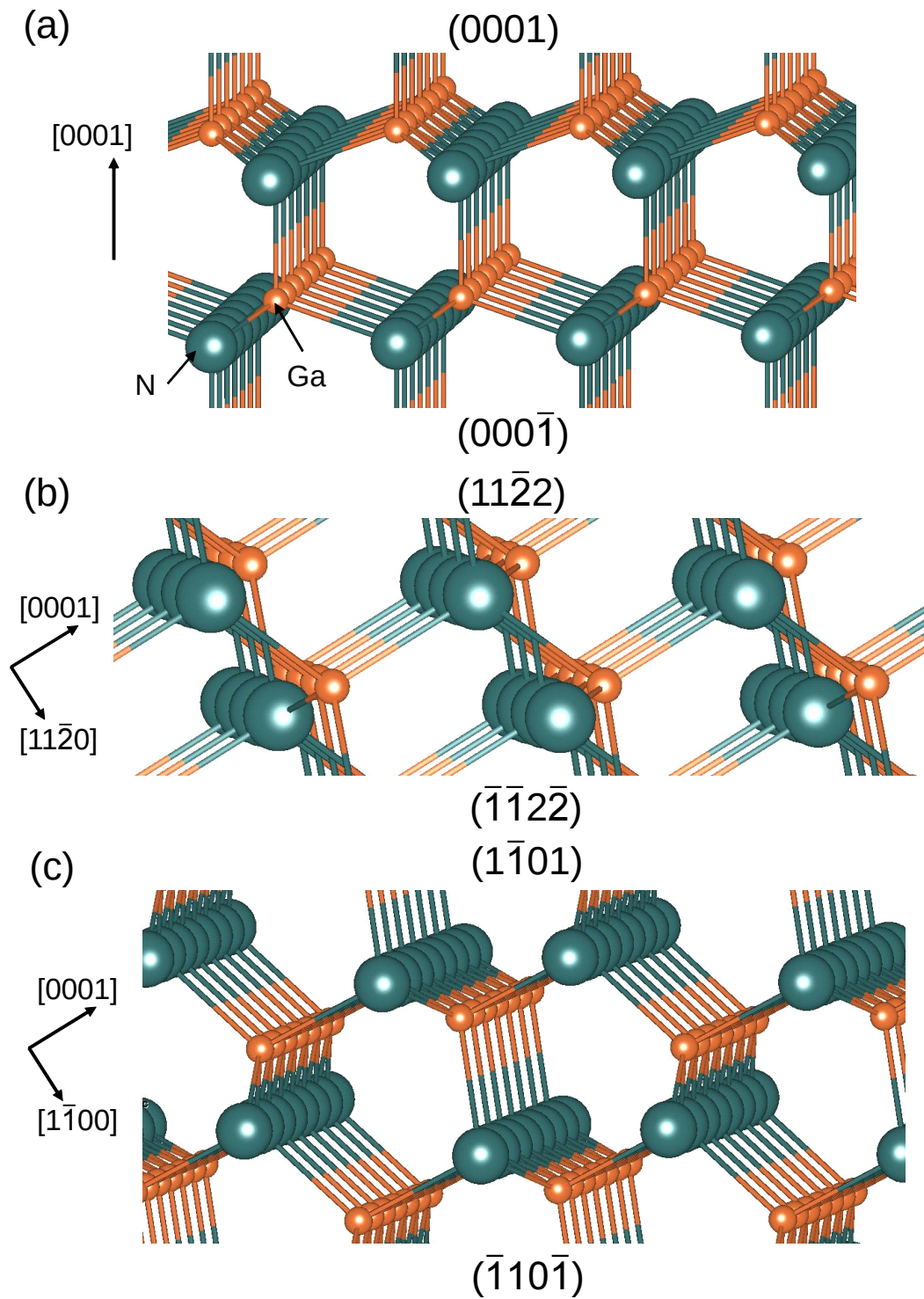


Figure 2.14: GaN structure schematics of (a) $\pm(0001)$, (b) $\pm(11\bar{2}\bar{2})$, and (c) $\pm(1\bar{1}01)$ ideal atomic arrangement.

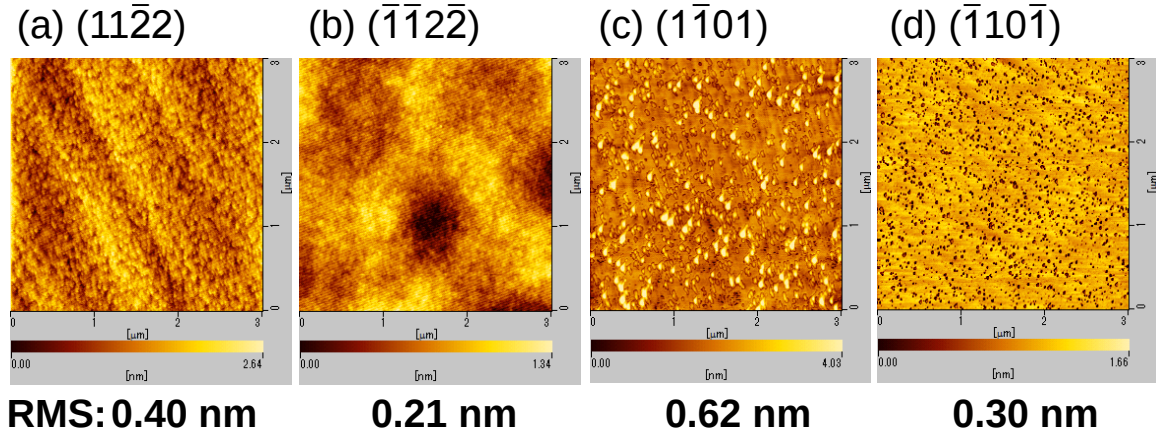


Figure 2.15: Surface morphologies of GaN homoepitaxial layers grown on (a) $(11\bar{2}2)$, (b) $(\bar{1}\bar{1}2\bar{2})$, (c) $(1\bar{1}01)$, and (d) $(\bar{1}\bar{1}0\bar{1})$ GaN substrates. All samples were grown under the same growth condition.

of conditions [25, 27]. These discussions suggested that the Ga adatom on $(\bar{1}\bar{1}2\bar{2})$ [or $(\bar{1}\bar{1}0\bar{1})$] migrate more actively than that on $(11\bar{2}2)$ [or $(1\bar{1}01)$], which is consistent with the trends observed in the 3D GaN on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$.

Although it is challenging to elucidate adatom behaviors on the growth surfaces directly, surface morphology observations of GaN films give us an insight into the adatom diffusion. This is because, the poor adatom mobility often leads to rough surface morphology caused by the high nucleation probability. Therefore, homoepitaxial GaN on $\pm(11\bar{2}2)$ and $\pm(1\bar{1}01)$ were fabricated, and the surface morphologies were confirmed by AFM measurements. The growth conditions were essentially the same as Fig. 3.2. Figure 2.15 summarizes the results. Comparing $\pm(11\bar{2}2)$ GaN [Figs. 2.15(a) and 2.15(b)], the $(\bar{1}\bar{1}2\bar{2})$ surface provides the smaller room mean square (RMS) roughness. In addition, Nishinaka reported that $(\bar{1}\bar{1}2\bar{2})$ GaN shows smooth surfaces under the wide range of the growth conditions, whereas the growth condition window providing smooth $(11\bar{2}2)$ GaN surface is very narrow [31]. Regarding $\pm(1\bar{1}01)$ GaN layers [Fig. 2.15(c) and 2.15(d)], the $(\bar{1}\bar{1}0\bar{1})$ surface provides the smaller RMS roughness. These comparisons experimentally indicate the active adatom migration on $(\bar{1}\bar{1}2\bar{2})$ and $(\bar{1}\bar{1}0\bar{1})$, corresponding to the top and inclined facets of the 3D GaN on $(\bar{1}\bar{1}2\bar{2})$. Therefore, the distinct growth evolutions between the 3D GaN on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ could be attributed to the adatom diffusivity on each crystallographic plane.

2.6 Summary

After introducing sample preparation procedures and MOVPE growth system, 3D GaN growth on $\{11\bar{2}2\}$ GaN substrates and the growth evolution were investigated. To explore favorable 3D GaN structures, several parameters such as substrate orientations $(11\bar{2}2)$ or $(\bar{1}\bar{1}2\bar{2})$, stripe directions, and mask geometries were prepared. The cross-sectional SEM observations clarified that 3D GaN on both the $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ planes with the same stripe mask pattern along $[\bar{1}\bar{1}23]$ are composed of semipolar and/or non-polar planes without (0001) polar plane, and seem to be promising for polychromatic emissions. Then, the growth processes of both the 3D GaN on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ were investigated by periodically inserted AlGaIn markers. The distinct growth evolutions were observed, and the analyses indicated active inter-facet migration during the 3D GaN on $(\bar{1}\bar{1}2\bar{2})$, which drastically affected the 3D shape evolution. The distinct growth evolutions between the 3D GaN on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ were considered to be originated from the surface atomic configurations affecting adatom migration.

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Chapter 3

3D Faceted InGaN QWs on $\{11\bar{2}2\}$ GaN Substrates

3.1 Introduction

In chapter 2, fabrication conditions to obtain polar-plane-free faceted 3D GaN structures on $\{11\bar{2}2\}$ GaN substrates were established. Both $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ surfaces with the particular stripe mask geometry provided such promising 3D GaN structures. Next challenge is to explore 3D faceted InGaN QWs favorable for radiative polychromatic emitters. As an introduction, expectations and concerns in this subject are briefly pointed out.

As mentioned in Sec. 2.1, polychromatic emission characteristics of 3D faceted InGaN QWs are introduced by the inter-facet distribution of In compositions and InGaN

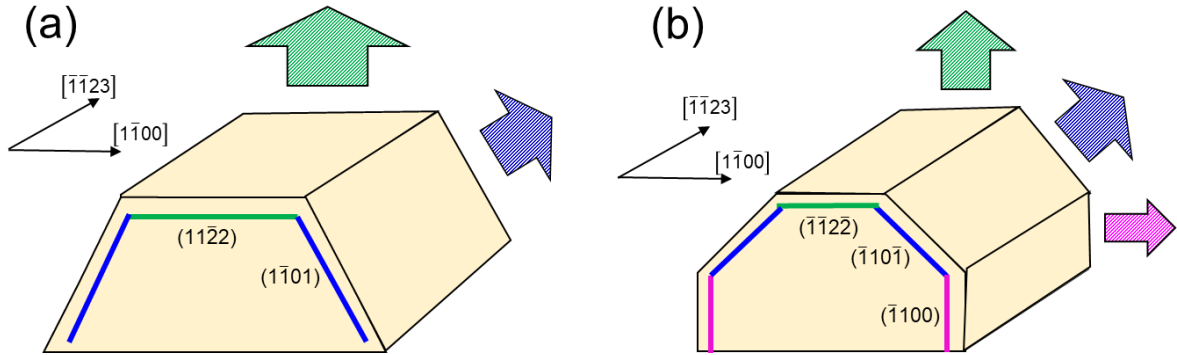


Figure 3.1: Schematic illustrations of expected emission characteristics of 3D faceted InGaN QWs on (a) $(11\bar{2}2)$ and (b) $(\bar{1}\bar{1}2\bar{2})$.

thicknesses. In the conventional 3D QWs on (0001), the top (0001) facet has the higher In composition and wider InGaN thickness than those of the inclined $\{11\bar{2}2\}$ and/or $\{1\bar{1}01\}$ facets [1–7], which is well accounted for by the inter-facet surface diffusion of In and Ga adatoms from the inclined to top facets [8,9]. Based on this consideration, similar emission wavelength distributions of the 3D InGaN QWs on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ can be expected as shown in Fig. 3.1. In both structures, the top facets emit longer wavelengths such as green, and the inclined and side facets emit shorter wavelengths like blue or violet.

Besides the inter-facet emission wavelength distributions, attention should also be directed to the material qualities. Several research groups have already reported polychromatic emissions from the 3D InGaN QWs on regrown GaN microstructures [1–7]. These structures, however, were fabricated on (0001) GaN templates. Regrown 3D GaN structures on (0001) have been extensively studied for high-quality GaN substrates, and effective reduction of threading dislocations was confirmed. On the other hand, in the case of $\{11\bar{2}2\}$ plane, Ni *et al.* found that basal-plane stacking faults (BSFs), which is originated from the heteroepitaxially-grown $(11\bar{2}2)$ GaN templates on *m*-plane sapphire substrates, propagate the overgrown 3D GaN structures [10]. The troublesome point is that BSFs usually work as nonradiative centers and significantly degrade the performance of the overgrown InGaN QWs. $\{11\bar{2}2\}$ GaN substrates used in this study are without BSFs, and thus the overgrown 3D GaN structures may not include BSFs. However, since little is known about the 3D GaN growth on semipolar planes, we should concern about that.

In this chapter, we demonstrate that polar-plane-free faceted 3D InGaN QWs on $(\bar{1}\bar{1}2\bar{2})$ exhibit facet-dependent polychromatic emission, whereas those on $(11\bar{2}2)$ shows monochromatic emission. Then, the local QW structural characterizations suggest that crystallographic orientation dependence of In incorporation affects inter-facet In adatom migrations, which results in distinct In composition distributions among 3D QWs on (0001), $(11\bar{2}2)$, and $(\bar{1}\bar{1}2\bar{2})$. Time-resolved photoluminescence (PL) spectroscopy reveals that 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ show shorter radiative recombination lifetimes due to the weakened piezoelectric fields. Finally, pastel color syntheses based on the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ are performed under photoexcitation condition.

3.2 InGaN QW Growth

InGaN/GaN QWs were subsequently grown on the 3D-GaN structures on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ with the mask geometry of fill factor (0.75). Figure 3.2 shows (a) structure

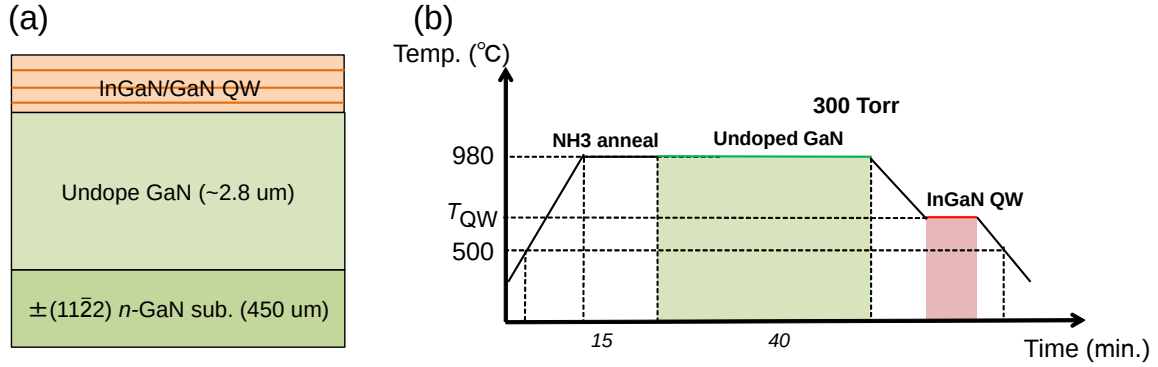


Figure 3.2: (a) A schematic structure on planar surface and (b) a temperature profile of 3D InGaN QWs on $(11\bar{2}\bar{2})$ and $(\bar{1}\bar{1}2\bar{2})$.

design on planar surface and (b) temperature profile used in this study. The growth condition of the 3D GaN structures was the same as Sec. 2.3. The growth temperature of InGaN QWs was 820 °C using N_2 as the carrier gas. Trimethylgallium (TMGa) and trimethylindium (TMIn) and NH_3 were used. The molar flow ratio between group III precursors (TMGa + TMIn) and NH_3 was 3000 for InGaN well. In/III ratio was 0.7. The designed InGaN well and GaN barrier thicknesses were ~ 3 nm and ~ 10 nm on a planar surface, respectively. The reactor pressure was set to 300 Torr. This growth condition was optimized in advance for planar $(11\bar{2}\bar{2})$ InGaN QWs in our laboratory.

3.3 Steady-State Optical Properties

The steady-state optical properties were assessed macroscopically with PL spectroscopy at room temperature (RT). A schematic diagram of the experimental setup is viewed in Fig. 3.3. The excitation source was a He-Cd laser (325 nm) with an excitation density of ~ 31.8 W/cm², and the emission was detected by a 25 cm spectrometer. Figure 3.4 shows the results for 3D QWs on (a) $(11\bar{2}\bar{2})$ and (b) $(\bar{1}\bar{1}2\bar{2})$. For comparison, PL spectra from planar QWs simultaneously grown with the 3D QWs are also indicated. The 366 nm emission is due to the GaN band-edge transition. The emission wavelength of the $(11\bar{2}\bar{2})$ planar QW is longer than that of the $(\bar{1}\bar{1}2\bar{2})$ planar QW (480 nm versus 425 nm), which can be attributed to a higher In incorporation efficiency of the $(11\bar{2}\bar{2})$ plane [11]. Figure 3.4(a) shows that the emission peak from the 3D QWs on $(11\bar{2}\bar{2})$ is located at ~ 480 nm. However, Fig. 3.4(b) shows that the PL spectrum of the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ has two peaks at ~ 390 nm and ~ 480 nm. That is, the emission from the 3D QWs on $(11\bar{2}\bar{2})$ is monochromatic, whereas that from the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$

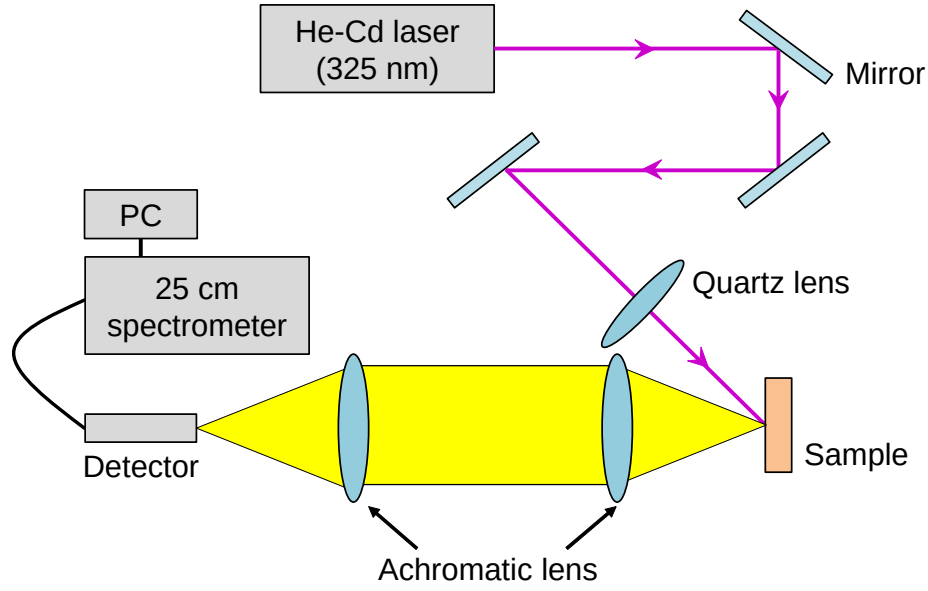
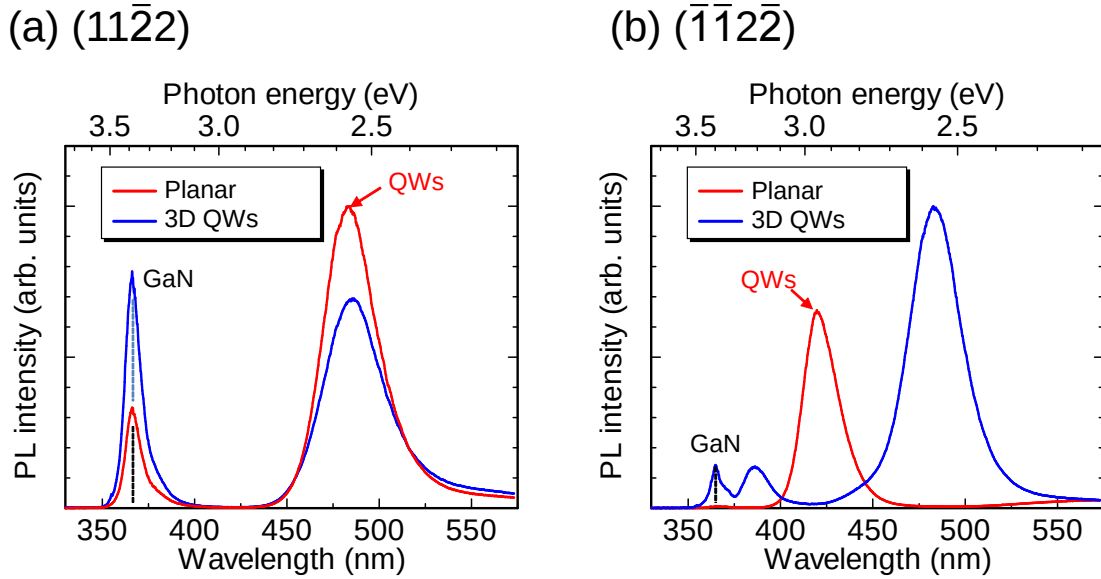


Figure 3.3: Schematic diagram of the PL measurement system.

Figure 3.4: RT-PL spectra from the 3D QWs grown on (a) $(11\bar{2}2)$ and (b) $(\bar{1}\bar{1}2\bar{2})$. The mask stripe is parallel to $[\bar{1}\bar{1}23]$ with an FF of 0.75. For reference, PL spectra from planar QWs simultaneously grown with 3D QWs are also given as the red lines.

is polychromatic. The reason for this difference is discussed below. Additionally, the intensity ratio of the 480 nm QW emission to the GaN band-edge emission of the 3D QWs on $(11\bar{2}2)$ is much higher than that on $(11\bar{2}\bar{2})$, which suggests that the former QWs have better optical properties owing to their superior structural qualities such as interface abruptness. The interface abruptness is supported by the cross-sectional SEM images shown in Fig. 2.7. In the following, we mainly focus on the detailed structural and optical properties of the 3D QWs on the $(\bar{1}\bar{1}2\bar{2})$ plane with the stripe mask parallel to $[\bar{1}\bar{1}23]$ with FF = 0.75.

Before describing detailed characterizations of 3D QWs on $(\bar{1}\bar{1}2\bar{2})$, let us discuss the origins of the poor interface abruptness of 3D QWs on $(11\bar{2}2)$. As mentioned in Chap. 2, Ueda and Nishinaka reported that optimal growth condition windows, which give atomically flat surfaces of planar $(11\bar{2}2)$ homoepitaxial GaN, are very narrow [12, 13]. Then, the local growth parameters at the window area in the selective area growth would be not optimum due to the precursor diffusion from the SiO_2 mask area, in addition to the gas phase diffusion.

The spatial distribution of the emission was evaluated by scanning electron microscopy (SEM) and cathodoluminescence (CL) spectroscopy at room temperature. For the 3D QWs on $(11\bar{2}2)$, emission peak wavelengths from the $(11\bar{2}2)$ and $(1\bar{1}01)$ facets located around 480 nm. Thus, the monochromatic emission characteristic of the 3D QWs on $(11\bar{2}2)$ observed in PL (Fig. 3.4(a)) well agrees with the uniform spatial distribution of the emission wavelength. In addition, monochromatic CL mapping at 480 nm represented a dark line along $[\bar{1}\bar{1}23]$ direction in the part of the top $(11\bar{2}2)$ facet, indicating defective region with considerable dislocation density or BSFs. The reason of the poor QW emission intensity found in Fig. 3.4(a) is partially attributed to the presence of this defective area. This finding suggests that selective area growth on semipolar plane may cause poor material qualities even under homoepitaxial growth. These results led us to conclude that the 3D QWs on $(11\bar{2}2)$ is unfavorable for highly efficient polychromatic emitters. Meanwhile, the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ show distinct results. Figure 3.5(a) displays an SEM bird's eye view of the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$. The cross-sectional image of the 3D structure is schematically illustrated on the right-hand side of Fig. 3.5(a). Figure 3.5(b) shows RT CL spectra acquired from the entire area of the SEM image and each facet designated in Fig. 3.5(a). The emission at 366 nm is from GaN. The CL emission peaks from the $(\bar{1}\bar{1}2\bar{2})$, $(\bar{1}10\bar{1})$, and $(\bar{1}100)$ facet QWs are located at approximately 440, 480, and 390 nm, respectively. Figures 3.5(c), 3.5(d), and 3.5(e) show monochromatic CL maps monitored at wavelengths of 390, 440, and 480 nm, respectively. The facet dependences of the emission peaks are confirmed.

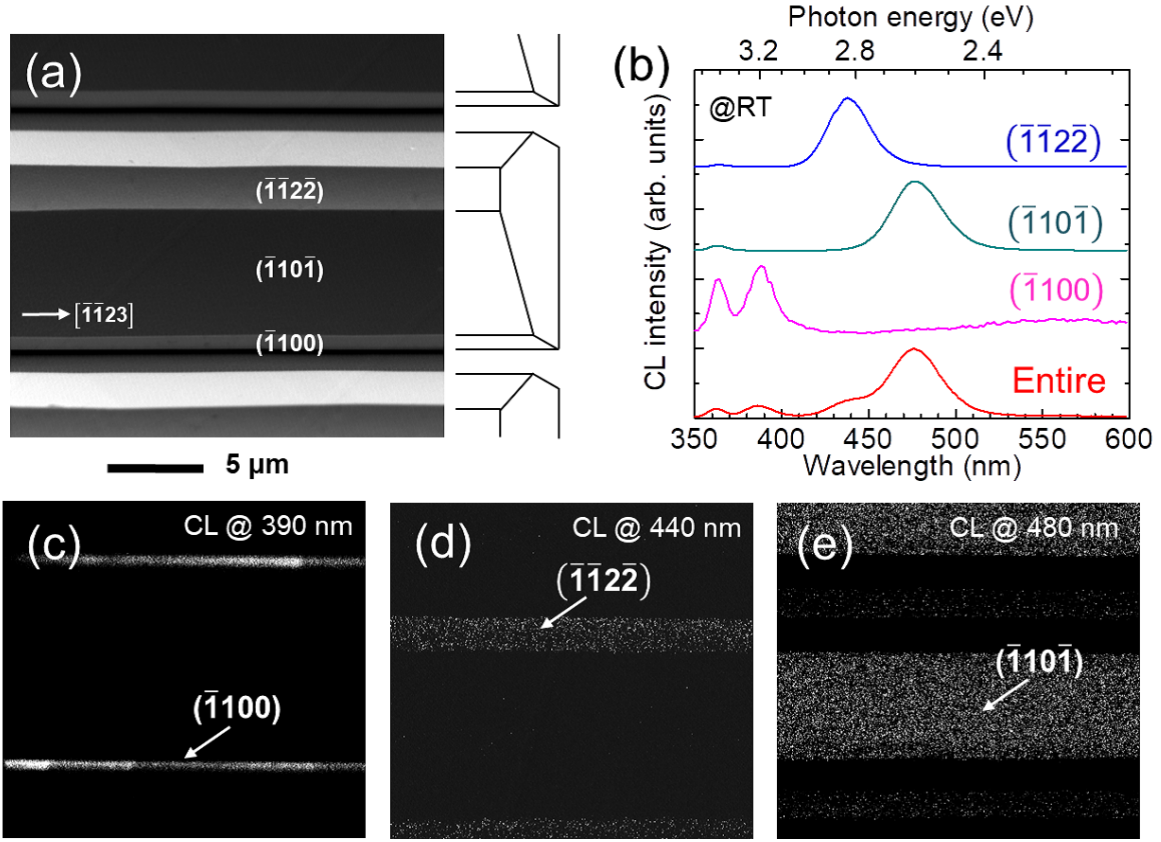


Figure 3.5: SEM bird's-eye view and schematic illustration of the 3D QWs on $(\bar{1}\bar{1}\bar{2}2)$. To see the $(\bar{1}\bar{1}00)$ nonpolar plane, the sample is tilted around the $[\bar{1}\bar{1}\bar{2}3]$ axis. (b) RT CL spectra acquired from the entire area and from the $(\bar{1}\bar{1}\bar{2}2)$, $(\bar{1}\bar{1}0\bar{1})$, and $(\bar{1}\bar{1}00)$ facets. Monochromatic CL mapping images taken at (c) 390 nm, (d) 440 nm, and (e) 480 nm.

In the CL spectrum from the entire region [Fig. 3.5(b)], the emissions from the $(\bar{1}\bar{1}\bar{2}2)$ and $(\bar{1}\bar{1}00)$ facet QWs are weaker than that from the $\{\bar{1}\bar{1}0\bar{1}\}$ facet QWs, most likely because of the smaller areas of the former facet QWs. Here, it should be noted that the 440 nm emission from the $(\bar{1}\bar{1}\bar{2}2)$ can be resolved in the CL spectrum of the entire area and, consistently, can be seen as a shoulder at 450 nm in the PL spectrum shown in Fig. 3.4(b). The relatively stronger $(\bar{1}\bar{1}\bar{2}2)$ emission in CL may be related to the experimental configuration; as a result of the sample rotation, the emission from one of the $\{\bar{1}\bar{1}0\bar{1}\}$ QWs is hardly detected, as demonstrated in Fig. Fig. 3.5(e), which strengthens the relative intensity of the $(\bar{1}\bar{1}\bar{2}2)$ emission.

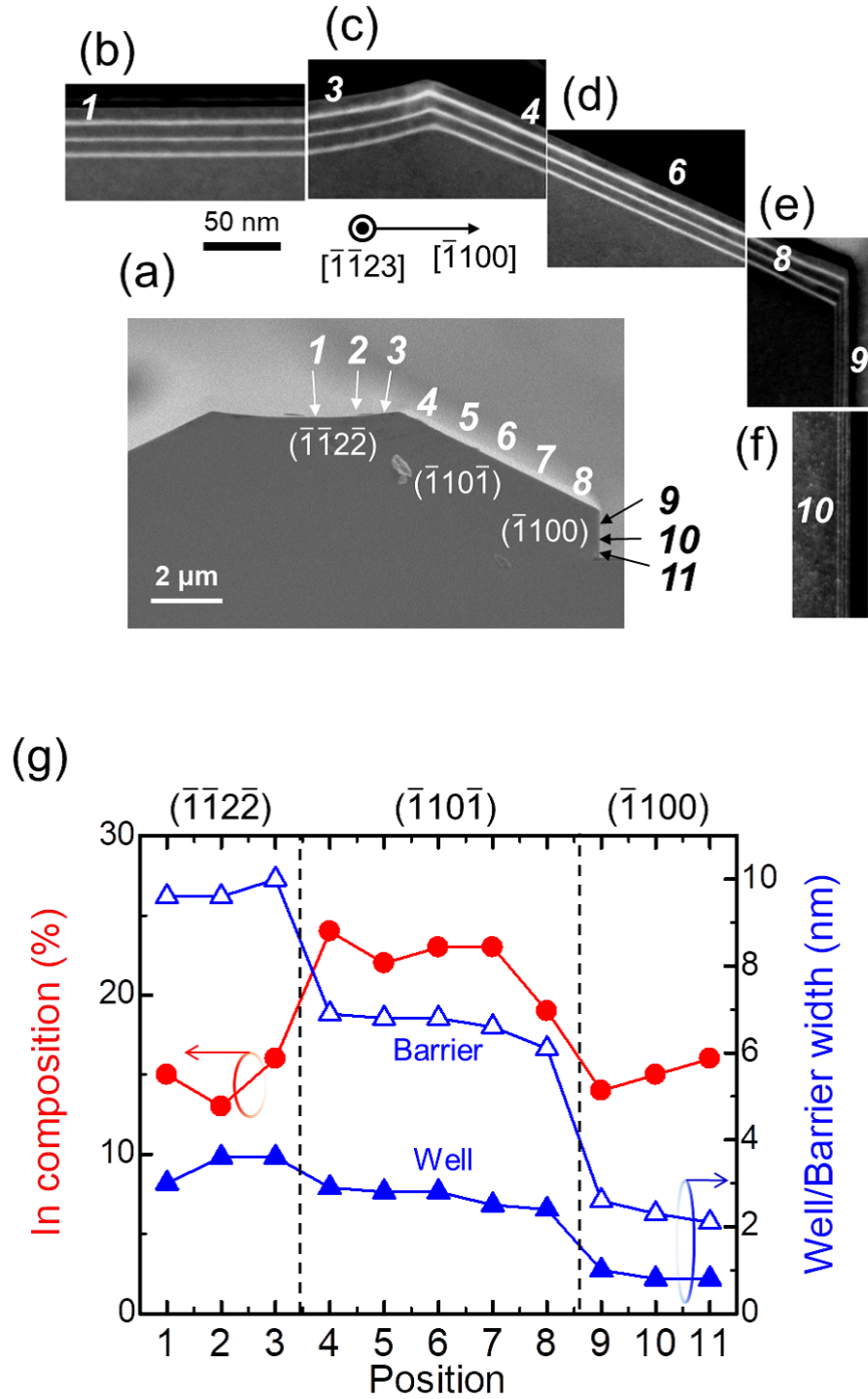


Figure 3.6: Structural characterization of the 3D QW on $(\bar{1}\bar{1}2\bar{2})$. (a) Cross-sectional SEM image. The numbers indicate the positions where HAADF-STEM, HR-TEM, and TEM-EDS measurements were performed. HAADF-STEM images at positions (b) 1, (c) 3-4, (d) 6, (e) 8-9, and (f) 10. The bright contrast is due to InGaN QWs. (g) Estimated local In composition, well width, and barrier width.

3.4 Structural Characterization

To clarify the origins of the facet dependent emission, the structural properties of the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ are investigated by transmission electron microscopy (TEM). Figure 3.6(a) shows a cross-sectional SEM image of the 3D QWs. Figures 3.6(b) to 3.6(f) show high-angle annular dark-field scanning TEM (HAADF-STEM) images taken at positions (b) 1, (c) 3-4, (d) 6, (e) 8-9, and (f) 10 indicated in Fig. 3.6(a). QW structures are clearly observed for all the facets, and it is found that the well and barrier widths depend on the location. The widths and compositions analyzed by high-resolution TEM (HR-TEM) and energy-dispersive X-ray spectroscopy (EDS) measurements under the STEM mode, respectively, are plotted in Fig. 3.6(g). The evaluation method of the In compositions by EDS elemental analyses can be found in Ref. [1]. The well and barrier widths increase with approaching from the sidewall $(\bar{1}100)$ to the top $(\bar{1}\bar{1}2\bar{2})$. On the other hand, the In composition varies in the order of $(\bar{1}10\bar{1}) > (\bar{1}\bar{1}2\bar{2}) \simeq (\bar{1}100)$. From the obtained In compositions, the calculated magnitudes of the electric fields due to the piezoelectric and spontaneous polarizations are ~ 0.39 MV/cm for $(\bar{1}\bar{1}2\bar{2})$, ~ 0.64 MV/cm for $(\bar{1}10\bar{1})$, and 0 MV/cm for $(\bar{1}100)$. Taking these quantities and the quantum confinement effect into account, the transition energies for $(\bar{1}\bar{1}2\bar{2})$, $\{\bar{1}10\bar{1}\}$, and $(\bar{1}100)$ facet QWs are estimated to be around 2.92, 2.66, and 3.25 eV, respectively. The PL and CL peak wavelengths in Figs. 3.4(b) and 3.5(b) are consistent with the calculations. (Small differences between the emission and transition energies may be due to the Stokes shift and exciton binding energy.) It is noteworthy that the emission wavelength from the $(\bar{1}\bar{1}2\bar{2})$ facet QW with the widest well width is shorter than that from the $(\bar{1}10\bar{1})$ facet QW with the highest In composition. This leads us to conclude that the polychromatic emission property shown in Figs. 3.4(b) and 3.5(b) is well accounted for by the facet-dependent In composition.

3.5 Growth Processes of 3D QWs on $\{11\bar{2}2\}$

We will move to discussions on the growth mechanisms of 3D faceted InGaN QWs on $(\bar{1}\bar{1}2\bar{2})$ GaN substrates. The structure is composed of $(\bar{1}\bar{1}2\bar{2})$, $\{\bar{1}10\bar{1}\}$, and $\{\bar{1}100\}$ planes, and the cross-sectional shape is similar to that of 3D QWs on (0001). However, the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ and (0001) show quite different inter-facet distribution of emission wavelength: for the 3D QWs on (0001), the longer wavelength emission relies on the top (0001) facet, whereas that relies on the inclined $\{\bar{1}10\bar{1}\}$ facets for those on $(\bar{1}\bar{1}2\bar{2})$. Figure 3.7 schematically illustrates the difference. In the case of the 3D QWs

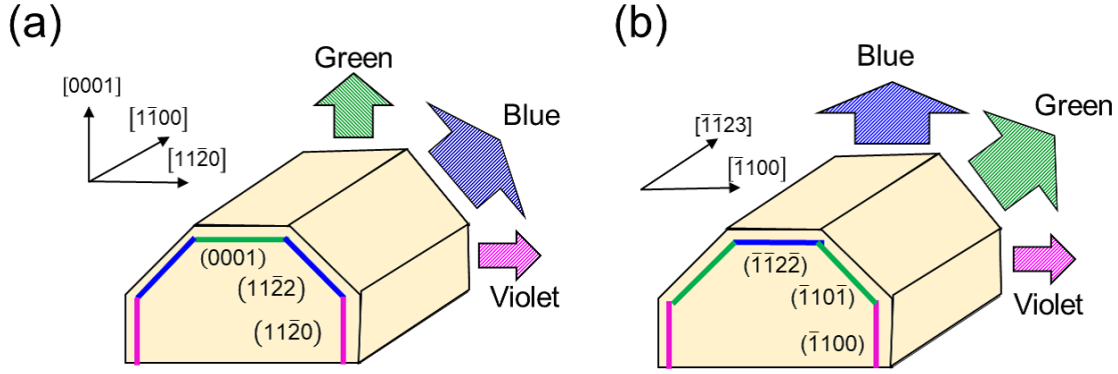


Figure 3.7: Schematic illustrations of emission characteristics of 3D QWs on (a) (0001) and (b) $(\bar{1}\bar{1}2\bar{2})$.

on (0001), the facet-dependence of the emission wavelength has been attributed to active In adatom migration from the inclined $\{11\bar{2}2\}$ to top (0001) facets [9]. Because both 3D QWs have similar trapezoidal cross sections, the observed difference indicates the presence of an unknown factor in addition to adatom migration. In this study, we discuss the growth mechanism that determines the inter-facet variation of the emission properties, comparing the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ with those on (0001). We demonstrate that the In incorporation efficiency into each facet plays an important role. Based on the clarified dynamics, origins of monochromatic emission property of the 3D QWs on $(11\bar{2}2)$ are also discussed. The findings are indispensable for structure designs and emission spectra control.

3.5.1 Local QW structural properties

Figure 3.8 shows cross-sectional SEM images and local QW structural properties of the 3D QWs on (0001) [(a) and (b)] and $(\bar{1}\bar{1}2\bar{2})$ [(c) and (d)]. In Figs. 3.8(b) and 3.8(d), open triangles are effective GaN and InN widths derived from the local InGaIn thickness and In composition. It should be noted that the well width and In composition in Fig. 3.8(b) were estimated by SEM and EDS equipped to the SEM; for this purpose, InGaIn thick films and InGaIn/GaN 200 period superlattices were grown on the 3D GaN structures because of the experimental resolution larger than the dimensions of QWs. On the other hand, those in 3.8(d) were estimated by TEM and EDS equipped to the TEM. We confirmed that both SEM + EDS and TEM + EDS provide essentially the same results. The detailed procedure and comparison can be found in Appendix A. Here, the top (0001) and $(\bar{1}\bar{1}2\bar{2})$ facets in Figs. 3.8(a) and 3.8(c) are flat and concave down,

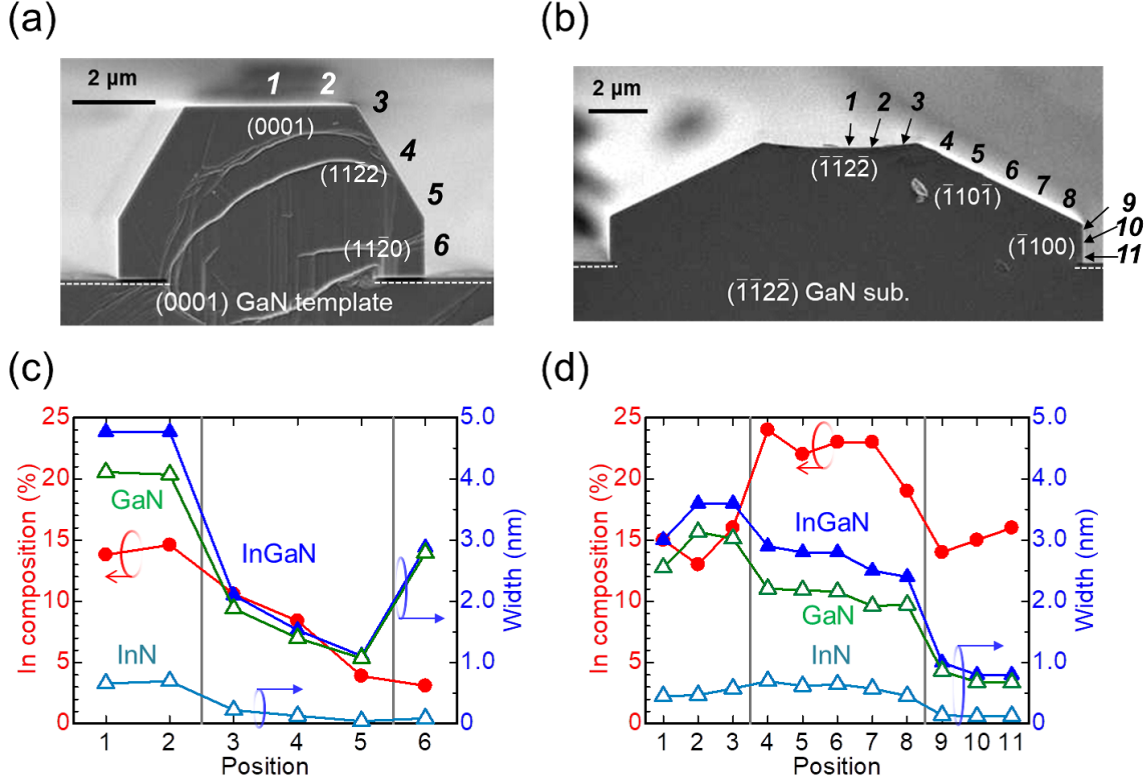


Figure 3.8: Cross-sectional SEM images and QW local structural properties of the 3D QWs on (0001) [(a) and (c)] and $(\bar{1}\bar{1}2\bar{2})$ [(b) and (d)], respectively.

respectively. The difference in the surface diffusion of adatoms between the inclined and top facets would be one of the plausible factors, as discussed in Sec 2.5.

Generally, the inter-facet emission-wavelength distributions can be accounted for by the local In composition and well width. For the 3D QWs on (0001) [Figs. 3.8(a) and 3.8(b)], the top (0001) facet QW has the widest well thickness and the highest In composition, resulting in the longest wavelength emission. For the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$, the well thickness distribution is similar to that of the 3D QWs on (0001). However, the QW with the highest In composition is formed on the inclined $(\bar{1}10\bar{1})$ facet, and the In composition ($\sim 23\%$) is much higher than that ($\sim 15\%$) of the top $(\bar{1}\bar{1}2\bar{2})$ facet. As a result, the longest-wavelength emission relies on the inclined $(\bar{1}10\bar{1})$ facet, which is also supported by the transition energy calculations in Sec. 3.4.

To further discuss the inter-facet variation of In compositions, the effective GaN and InN thickness distributions in Figs. 3.8(b) and 3.8(d) are compared. In general, a film thickness d should be proportional to a growth rate v . Moreover, the growth rate can

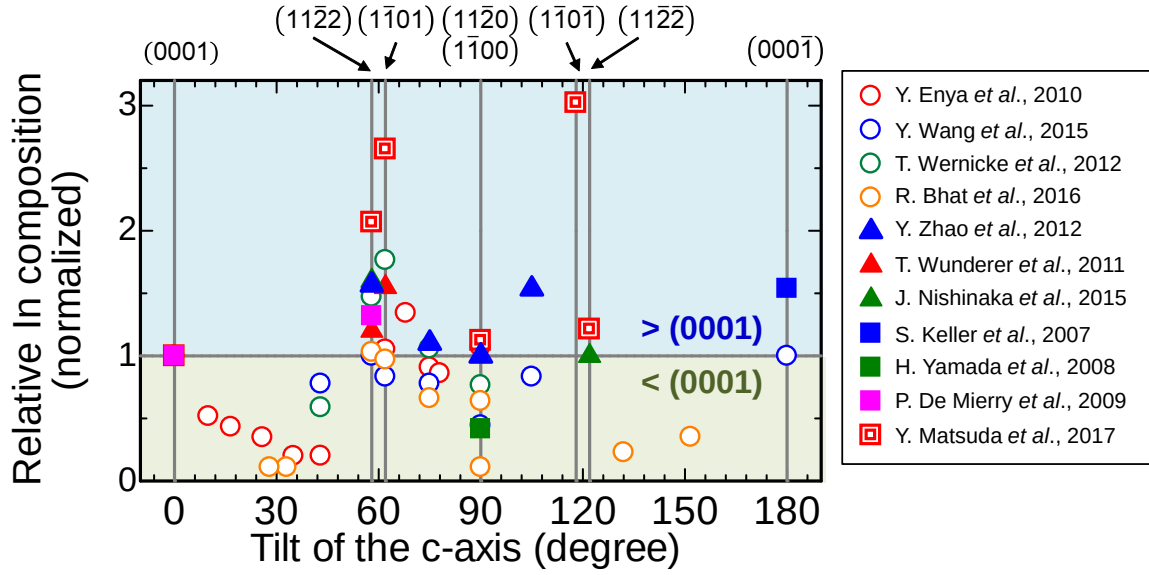


Figure 3.9: Normalized In composition with respect to the (0001) plane. All the data are cited from [6, 11, 14–22].

be approximated as $v \propto kJ$, where k and J are incorporation efficiency and the sum of fluxes of atoms from the gas phase and adatoms migrating on the surface, respectively. Thus, higher k and/or J result in a thicker film, as already mentioned in Sec. 2.5. For the 3D QWs on (0001) [Fig. 3.8(b)], GaN and InN thicknesses increase from the inclined $(11\bar{2}2)$ to the top (0001) facets. This was attributed to the active Ga and In adatom migration from the inclined $(11\bar{2}2)$ to the top (0001) facets. Furthermore, more active In adatom migration, which can read from the thickness ratio between the (0001) and $(11\bar{2}2)$ facets, leads to the In-rich QWs on the top (0001) facet. (In adatoms are easier to evaporate than Ga adatoms due to the higher vapor pressure.) That is, the GaN and InN thicknesses in the 3D QWs on (0001) are chiefly determined by the mass transport (J). Meanwhile, for the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ [Fig. 3.8(d)], the GaN thickness distribution exhibits a trend similar to that of the 3D QWs on (0001), which means that the GaN thickness also depends on the mass transport. However, the InN thickness slightly decreases from the inclined $(\bar{1}10\bar{1})$ to the top $(\bar{1}\bar{1}2\bar{2})$ facets, which suggests that the InN thickness in the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ depends not only on the mass transport (J) but also on the facet-dependent In incorporation efficiency (k).

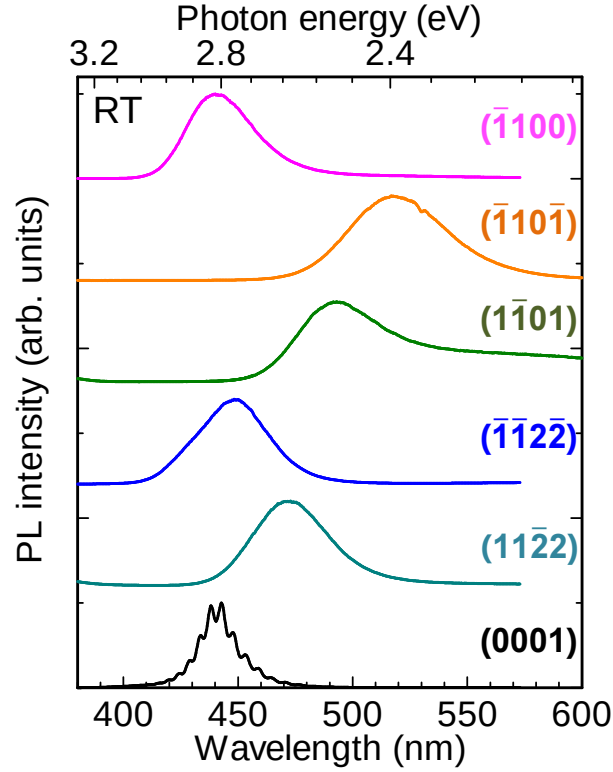


Figure 3.10: Typical PL spectra from (0001), $\pm(11\bar{2}2)$, $\pm(1\bar{1}01)$, and $(\bar{1}100)$ InGaN QWs at RT.

3.5.2 Crystallographic orientation dependence of In incorporation

Multiple papers reported In compositions of InGaN QWs grown on differently oriented substrates [6, 11, 14–22]. Figure 3.9 summarizes the orientation dependence of experimental In composition normalized with respect to that of the (0001) plane. Most of the reports show that the $(11\bar{2}2)$ and $(1\bar{1}01)$ planes exhibit higher In compositions than the (0001) plane. For example, T. Wernicke *et al.* reported the following order: $(1\bar{1}01) > (11\bar{2}2) > (0001) > (20\bar{2}1) > (1\bar{1}01)$ [19]. However, there is no direct comparison among (0001), $\pm(11\bar{2}2)$, and $\pm(1\bar{1}01)$ planar QWs. Therefore, we fabricated those QWs on respective GaN substrates simultaneously under the same growth condition as the 3D QWs, and we estimated QW structural parameters by XRD.

Figure 3.10 displays typical PL spectra from the fabricated (0001), $\pm(11\bar{2}2)$, $\pm(1\bar{1}01)$, and $(\bar{1}100)$ planar QWs at RT. The PL system was the same as Fig. 3.3. The following order of the emission wavelength is indicated: $(\bar{1}101) > (1\bar{1}01) > (11\bar{2}2) > (\bar{1}1\bar{2}2) \simeq$

Table 3.1: Estimated well width (WW), barrier width (BW), and In composition (In) of simultaneously grown planar QWs.

Plane	WW (nm)	BW (nm)	In (%)
(0001)	3.4	9.6	8.9
(11 $\bar{2}2$)	3.7	10.4	18.4
($\bar{1}\bar{1}2\bar{2}$)	3.6	10.1	10.8
(1 $\bar{1}01$)	2.7	9.3	23.6
($\bar{1}10\bar{1}$)	2.5	10.6	26.9
($\bar{1}100$)	7.2	7.2	10.0

($\bar{1}100$) $\simeq$ (0001). Table 3.1 summarizes the structural parameters. The In compositions vary in the order of 26.9% for ($\bar{1}10\bar{1}$) > 23.6 % for (1 $\bar{1}01$) > 18.4% for (11 $\bar{2}2$) > 10.8% for ($\bar{1}\bar{1}2\bar{2}$) $\simeq$ 10.0% for ($\bar{1}100$) $\simeq$ 8.9% for (0001) (These results are also plotted in Fig. 3.9). Here, let us note current status of theoretical investigations for the orientation dependence of In incorporation. Although no clear conclusion has yet to be elucidated, orientation dependence of In incorporation has been explained in terms of several physical origins. Durnev *et al.* explained those dependencies using calculated strain energy, which depends on substrate orientation [23]. The author predicted that the strain reduction leads to a higher In incorporation on both nonpolar and semipolar planes. Experimentally, semipolar (11 $\bar{2}2$) and (1 $\bar{1}01$) planes exactly show higher In compositions, but nonpolar (1100) and (11 $\bar{2}0$) planes have similar or smaller In compositions compare to (0001) plane, as shown in Fig. 3.9. Alternatively, Yayama *et al.* constructed a theoretical model that explains the role of the surface N-H layer in In incorporation based on first-principles calculations [24]. According to the surface phase diagrams, the growth surfaces are terminated by NH_i ($i=0, 1, 2, 3$) molecules for all orientations under the typical InGaN growth condition. The H-terminated N-layer, which covers group-III atoms, is assumed to maintain a high In composition, and the effective enthalpy change for N-H layer decomposition to NH_i molecules ($\Delta H_{\text{dec}}^{\text{eff}}$) was calculated using the enthalpy change for the N-H layer decomposition (ΔH_{dec}) and N-coverage (θ). Namely, a larger $\Delta H_{\text{dec}}^{\text{eff}}$ means a higher In composition. The calculated results well explain the dependence of $\pm(0001)$, $\pm(11\bar{2}2)$, (1 $\bar{1}01$), and ($\bar{1}\bar{1}00$) planes. Comparing with our results in Tab. 3.1, the only exception is ($\bar{1}10\bar{1}$) plane: they theoretically predicted ($\bar{1}10\bar{1}$) $\simeq$ ($\bar{1}\bar{1}2\bar{2}$), but we experimentally observed that ($\bar{1}10\bar{1}$) plane

has the highest In composition among the examined orientations. Therefore, the effect of surface reconstruction terminated by N-H molecules could be one of the plausible factors determining In incorporation, and the higher In composition of the $(\bar{1}10\bar{1})$ plane might be derived from the different surface reconstruction from their calculations.

Here, again, the local QW structural parameters of the 3D QWs on (0001) and $(\bar{1}\bar{1}2\bar{2})$ are discussed, taking into account the crystallographic orientation dependence of In incorporation. For the 3D QWs on (0001), the top (0001) facet QWs have a higher In composition than the inclined $(11\bar{2}2)$ facet QWs, in spite of the lower In incorporation efficiency. This observation is consistent with the active In adatom migration from the inclined $(11\bar{2}2)$ to the top (0001) facets, which suggests that the higher In incorporation efficiency of the $(11\bar{2}2)$ plane is hindered by such active In adatom migration. On the other hand, the In compositions of the $(\bar{1}\bar{1}2\bar{2})$ and $(\bar{1}10\bar{1})$ facet QWs in the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ have a tendency similar to those of each planar QWs. Here, it should be noted that the $(\bar{1}10\bar{1})$ plane shows a significantly higher In composition, which is presumed to promote efficient incorporation of In adatoms and suppress the In adatom migration to the top $(\bar{1}\bar{1}2\bar{2})$ facet. As a result, the InN thickness of the inclined $(\bar{1}10\bar{1})$ facet becomes slightly thicker than that of the top $(\bar{1}\bar{1}2\bar{2})$ facet [Fig. 3.8(d)].

3.5.3 Diffusion equation analyses

To support the above assertion, diffusion equation analyses were performed. Figure 3.11(a) shows a cross-sectional SEM image and the definitions of the coordinates. The 3D structure was grown on a mask pattern with a larger window width than that in Fig. 3.8(c) to appropriately estimate the adatom diffusion length on the top $(\bar{1}\bar{1}2\bar{2})$ facet. The local InGaN thickness and In compositions were estimated by SEM and EDS equipped to the SEM in the same manner as Fig. 3.8(b). The estimated local thickness of GaN and InN on both $(\bar{1}\bar{1}2\bar{2})$ and $(\bar{1}10\bar{1})$ facets were plotted in Figs. 3.11(b) and 3.11(c), respectively. These thickness distributions were fitted based on the diffusion equation. The detailed procedure can be found in Ref. [9], but the thickness distributions were fitted by the following formula derived from the diffusion equation,

$$N_{adatom} = A_1 \exp\left(\frac{x}{\lambda_{adatom}}\right) + A_2 \exp\left(-\frac{x}{\lambda_{adatom}}\right) + B, \quad (3.1)$$

where N is local density of adatom, $adatom$ is Ga or In, A_1 and A_2 are amplitudes, and B is the constant term. For Ga, the estimated surface diffusion lengths, $\lambda_{\text{Ga}}^{(\bar{1}\bar{1}2\bar{2})}$ and $\lambda_{\text{Ga}}^{(\bar{1}10\bar{1})}$, are calculated to be 6.2 and 4.2 μm , respectively. For In, $\lambda_{\text{In}}^{(\bar{1}\bar{1}2\bar{2})}$ and $\lambda_{\text{In}}^{(\bar{1}10\bar{1})}$ are

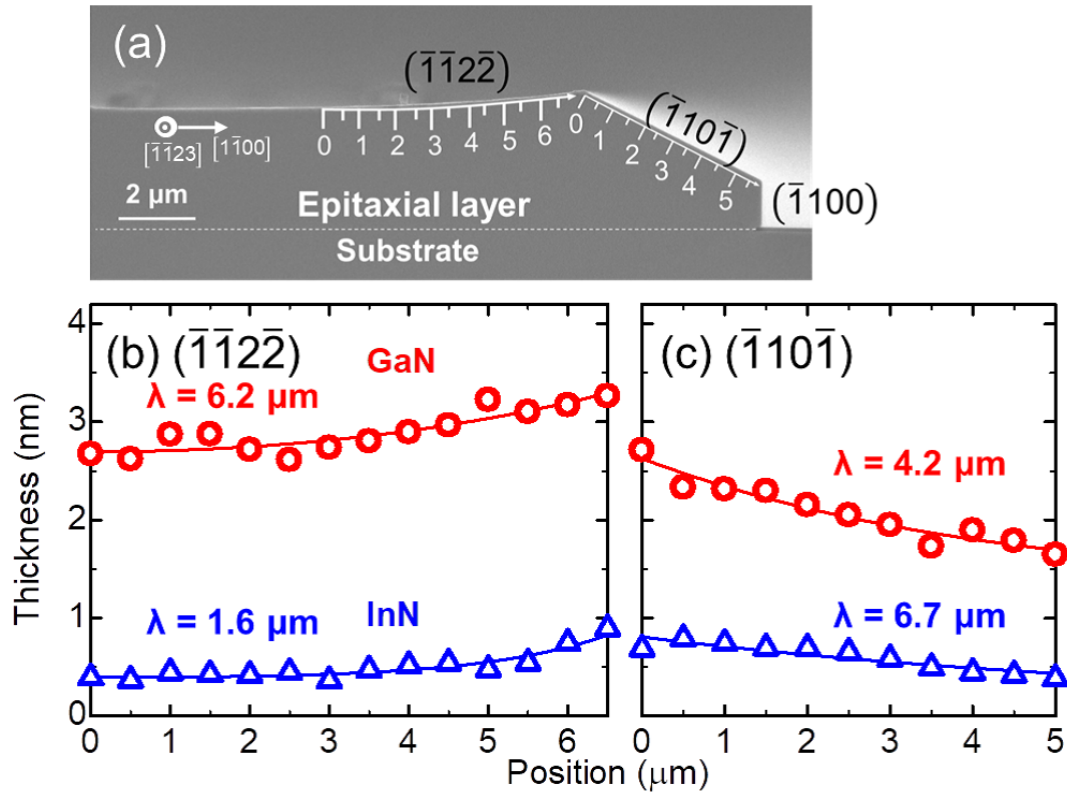


Figure 3.11: (a) Cross-sectional SEM image. Local GaN and InN thicknesses on (b) $(\bar{1}\bar{1}2\bar{2})$ and (c) $(\bar{1}10\bar{1})$ facets at positions designated in (a).

1.6 and 6.7 μm , respectively.

Note that higher growth temperatures generally shorten diffusion lengths by promoting evaporation, while activating migration. Therefore, a shorter diffusion length suggests more active migration. For the 3D QWs on (0001), the Ga diffusion length on the top (0001) facet was 5.2 μm , while that on the inclined $(11\bar{2}2)$ facet was 3.1 μm [9]. This trend is quite similar to the present observation, which suggests that Ga adatoms behave similarly on both the 3D QWs. On the other hand, the In diffusion lengths show the opposite tendency between the 3D QWs on (0001) and $(\bar{1}\bar{1}2\bar{2})$; on (0001), those were 2.7 μm on the top facet and 1.6 μm on the inclined facet [9], which suggests more drastic migration on the inclined $\{11\bar{2}2\}$ facets. Such active In adatom migration overcomes the higher In incorporation efficiency of the $(11\bar{2}2)$ plane, resulting in a lower In composition. On the other hand, for the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$, the much longer In diffusion length on the inclined $(\bar{1}10\bar{1})$ than that on the top $(\bar{1}\bar{1}2\bar{2})$ means passive In adatom migration on the inclined $(\bar{1}10\bar{1})$ facets most likely due to the higher In incorporation efficiency. These findings are consistent with the assertion mentioned

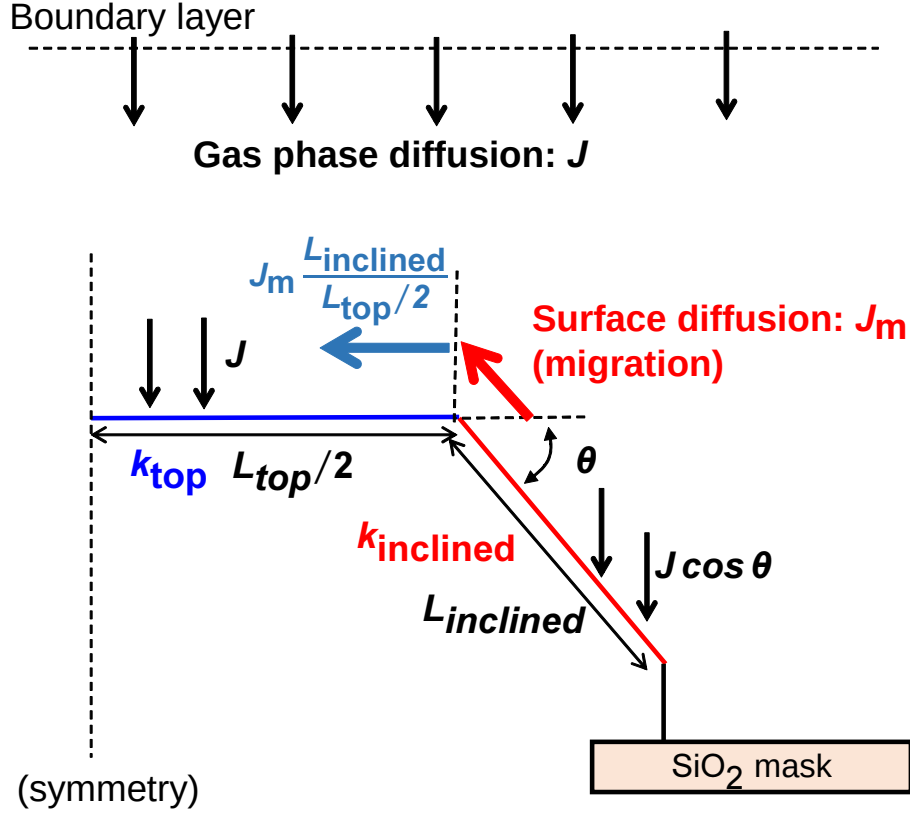


Figure 3.12: A model to simulate the growth dynamics on the 3D structure. The fluxes of atoms from the gas phase and adatoms migrating on the surface are indicated by the black and colored arrows, respectively.

above.

3.5.4 A model calculation of mass transport under the influence of In incorporation

In this section, we propose a simple model based on the one-dimensional inter-facet surface migration. In this model, the flux of adatoms migrating from the inclined to top facet J_m is assumed to be influenced by the atom incorporation efficiency into the solid phase. The atom incorporation efficiency of each crystallographic plane is described in Sec. 3.5.1. This is expanded from the model constructed in Sec. 2.5.2.

Figure 3.12 schematically shows this model, where parameters on the top and inclined planes are represented as subscripts, while those on the planar and facet QWs

Table 3.2: Effective GaN and InN thickness of the top and inclined facets in the 3D QWs on (0001) and $(\bar{1}\bar{1}2\bar{2})$. Those are the averaged values of the plots for the each facet in Figs. 3.8(c) and 3.8(d).

3D QWs	Plane	GaN (nm)	InN (nm)
on (0001)	Top (0001)	4.09	0.68
	Inclined $(11\bar{2}2)$	1.45	0.13
on $(\bar{1}\bar{1}2\bar{2})$	Top $(\bar{1}\bar{1}2\bar{2})$	2.90	0.50
	Inclined $(\bar{1}10\bar{1})$	2.08	0.60

are designated by superscripts. From the relationship $d \propto v \propto kJ$, we obtain

$$\frac{J_{\text{top}}^{\text{facet}}}{J_{\text{inclined}}^{\text{facet}}} = \frac{d_{\text{top}}^{\text{facet}}/k_{\text{top}}}{d_{\text{inclined}}^{\text{facet}}/k_{\text{inclined}}}. \quad (3.2)$$

The incorporation ratio, $k_{\text{top}}/k_{\text{inclined}}$, is equal to the film thickness ratio of planar QWs grown along the corresponding directions under the same condition, that is, $d_{\text{top}}^{\text{planar}}/d_{\text{inclined}}^{\text{planar}}$, because J is common for the planar QWs irrespective of the crystallographic orientations. Then, we have

$$\frac{J_{\text{top}}^{\text{facet}}}{J_{\text{inclined}}^{\text{facet}}} = \frac{d_{\text{top}}^{\text{facet}}/d_{\text{top}}^{\text{planar}}}{d_{\text{inclined}}^{\text{facet}}/d_{\text{inclined}}^{\text{planar}}}. \quad (3.3)$$

On the inclined facet, the flux from the gas phase becomes $J \cos \theta$ due to the geometrical configuration, where θ is the angle between the top and inclined facets. In addition, adatoms are assumed to migrate from the inclined to top facet with a flux of J_{m} . Thus, the total flux on the inclined facet is expressed as $J \cos \theta - J_{\text{m}}$. Noting that J_{m} (as well as J) is defined as an area density, the flux into the top facet can be expressed as $J + J_{\text{m}} \frac{L_{\text{inclined}}}{L_{\text{top}}/2}$, where L_{top} and L_{inclined} are the facet widths of the 3D structures. Therefore, we obtain

$$\frac{J + J_{\text{m}} \frac{L_{\text{inclined}}}{L_{\text{top}}/2}}{J \cos \theta - J_{\text{m}}} = \frac{d_{\text{top}}^{\text{facet}}/d_{\text{top}}^{\text{planar}}}{d_{\text{inclined}}^{\text{facet}}/d_{\text{inclined}}^{\text{planar}}}. \quad (3.4)$$

The assumptions in this model are as follows: (1) only the half of the top facet (width of $L_{\text{top}}/2$) is considered due to the structural symmetry, (2) J_{m} is constant on the entire area of the inclined facet, and (3) adatom diffusion to/from the side facet can be ignored

due to the small area.

For the planar QWs, we have experimentally determined the thickness ratios as $d_{(0001)}^{\text{planar}}/d_{(11\bar{2}2)}^{\text{planar}} = 1.02$ and 0.44 for GaN and InN, respectively, and similarly, $d_{(11\bar{2}2)}^{\text{planar}}/d_{(1\bar{1}01)}^{\text{planar}}$ is 1.75 for GaN and 0.58 for InN [22]. For the 3D structures, the thicknesses were evaluated from Figs. 3.8(b) and 3.8(d), as summarized in Tab. 3.2. Using these values with Eq. (3.4), the normalized flux J_m/J is calculated to be $+0.09$ for GaN and $+0.37$ for InN in the 3D QWs on (0001), which means that surface In adatoms more drastically migrate from the inclined to top facets than Ga adatoms. On the other hand, for the 3D QWs on $(1\bar{1}\bar{2}2)$, J_m/J is -0.09 for GaN and $+0.08$ for InN. It is noteworthy that J_m/J for InN in the 3D QWs on $(1\bar{1}\bar{2}2)$ is much smaller than that in 3D QWs on (0001), which indicates that less In adatoms migrate from the inclined to top facets. This trend well agrees with the above discussion. Although the surface diffusion direction of Ga adatoms in the 3D QWs on $(1\bar{1}\bar{2}2)$ is inconsistent with the diffusion equation analyses, J_m/J for GaN is relatively small, and it may be affected by the assumptions in our model.

3.5.5 3D QWs on $(11\bar{2}2)$ vs. $(1\bar{1}\bar{2}2)$

The aforementioned monochromatic emission property of the 3D QWs on $(11\bar{2}2)$ [Fig. 3.4(a)] can be also explained by inter-facet adatom migration and orientation dependence of In incorporation. Figure 3.13 compares cross-sectional SEM images and local QW structural properties of the 3D QWs on $(11\bar{2}2)$ [(a) and (c)] and $(1\bar{1}\bar{2}2)$ [(b) and (d)]. Effective GaN and InN widths were derived in the same manner in Sec. 3.5.1. It should be noted that the well width and In composition in Fig. 3.13(c) were estimated by SEM and EDS equipped to the SEM. In Fig. 3.13(c), the InGaN well width decreases from the top to inclined facets, similar with the 3D QWs on $(1\bar{1}\bar{2}2)$ [Fig. 3.13(d)] and also on (0001) [Fig. 3.8(c)]. On the other hand, the inclined $(1\bar{1}01)$ facet has slightly higher In composition than the top $(11\bar{2}2)$ facet. This distribution results in the monochromatic emission. Then, the effective GaN and InN thickness distributions indicate the growth dynamics. The GaN distribution exhibits the same trend as the 3D QWs on $(1\bar{1}\bar{2}2)$ and (0001), but the InN distribution is relatively uniform compared with the 3D QWs on (0001), indicating slightly suppressed In adatom migration on the inclined facet. Actually, the $(1\bar{1}01)$ planar QW has a slightly higher In incorporation efficiency than the $(11\bar{2}2)$ planar QW (Tab. 3.1), which is compensated for by the migration effect, eventually leading to a small inter-facet variation of the In composition.

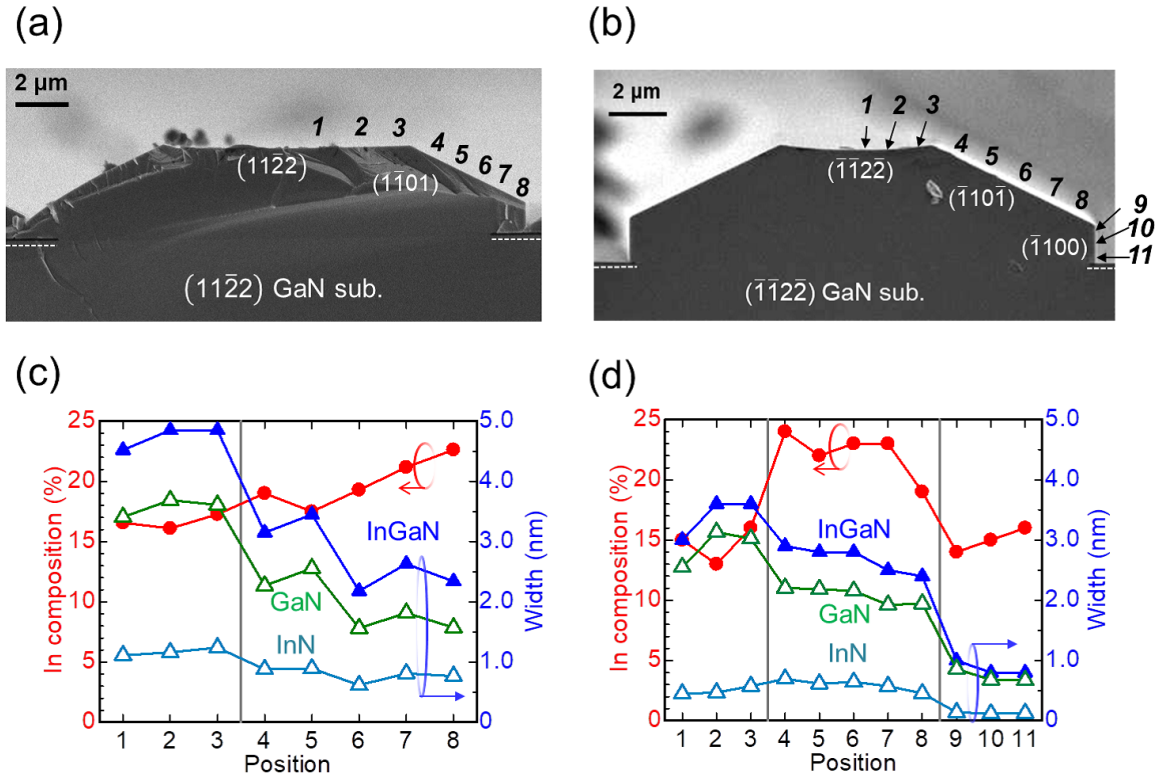


Figure 3.13: Cross-sectional SEM images and QW local structural properties of the 3D QWs on (0001) [(a) and (c)] and $(\bar{1}\bar{1}2\bar{2})$ [(b) and (d)], respectively.

Inter-facet variation of the In composition affected by In incorporation efficiencies is also found in Ref. [6]. The inversed pyramids composed of closely neighboring $\{1\bar{1}01\}$ and $\{11\bar{2}2\}$ facet QWs provide facet-dependent polychromatic emission; the emission wavelength of the $\{1\bar{1}01\}$ facet QWs is 20 nm longer than the $\{11\bar{2}2\}$ facet QWs. This tendency can be explained by a similar scenario based on the higher In incorporation efficiency of the $(1\bar{1}01)$ plane.

3.6 Spontaneous Emission Lifetime

The developed 3D QWs consist of semipolar $(\bar{1}\bar{1}2\bar{2})$ and $\{1\bar{1}0\bar{1}\}$ and nonpolar $\{1\bar{1}00\}$ planes. Due to the reduced internal electric fields on the semi- and non-polar planes, this 3D QW structure can avoid the problem caused by the polar (0001) plane. TPRL is a powerful tool to demonstrate the effect of the reduced internal electric fields. The carrier lifetimes measured at low temperature (LT), where the estimated lifetimes are chiefly

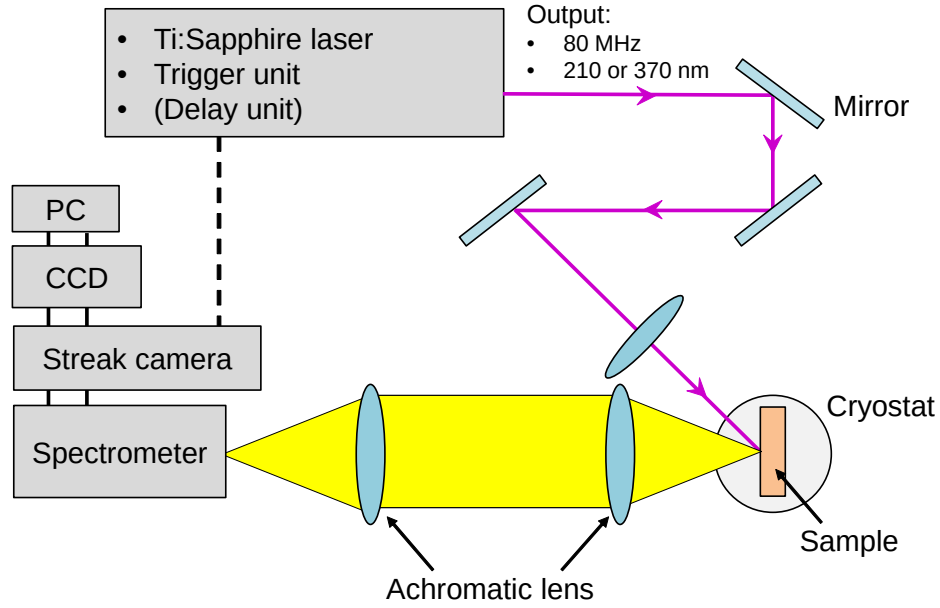
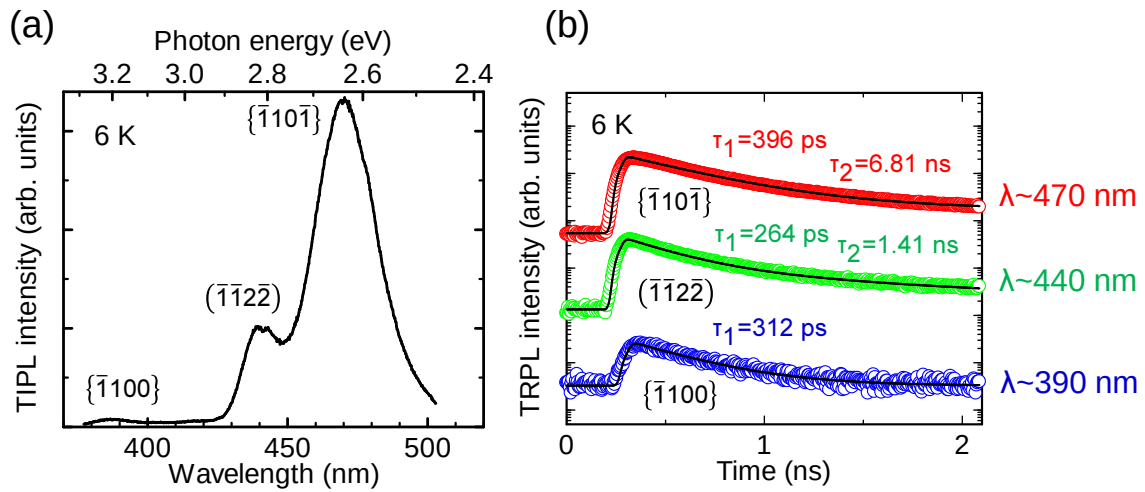


Figure 3.14: Schematic diagram of the TRPL measurement system.

Figure 3.15: (a) TIPL spectrum of the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$, and (b) PL decay curves from the $\{110\bar{1}\}$, $(\bar{1}\bar{1}2\bar{2})$, and $\{1100\}$ facets. The experiments were performed at ~ 6 K.

affected by the radiative recombination processes, are a good index of the strength of the internal electric fields; large internal electric fields separate electrons and holes inside the QWs, leading to the longer radiative recombination lifetimes. A number of studies has already demonstrated the faster radiative recombination lifetimes from the semi/nonpolar facet QWs of the 3D QWs on (0001) GaN templates or semi/nonpolar-based planar InGaN QWs [1, 25–34]. The common trends are as follows: (0001) polar QWs exhibit the radiative recombination lifetimes ranged from a few tens of ns to μ s order depending on the emission colors, whereas semi/nonpolar QWs have those of around a few hundred ps to a few tens of ns. Most of the previous reports are in the range of violet to green emission colors. Accordingly, the developed 3D QWs can also be expected to exhibit shorter lifetimes about a few ns or less.

Let us move to the experimental conditions in this study. A schematic diagram of the measurement system is viewed in Fig. 3.14, and the measurements were performed at ~ 6 K. The excitation source was a Ti:Al₂O₃ laser with a pulse repetition rate and width of 80 MHz and 2 ps, respectively. The output wavelength was set to 212 or 370 nm, and the excitation density was ~ 1.6 nJ/cm². From those quantities, the photogenerated carrier densities were estimated to be, at most, 2.7×10^{14} cm⁻³. Thus, the current experimental conditions can be regarded as weak excitation conditions, because such a carrier density barely affects internal electric fields [35].

Figure 3.15(a) displays a time-integrated PL (TIPL) spectrum from the 3D QWs on ($\bar{1}\bar{1}2\bar{2}$) excited at 212 nm wavelength. Figure 3.15(b) shows PL decay curves acquired at around 390, 440, and 470 nm. From the discussions in Sec. 3.4, the TIPL peaks at about 390, 440, and 470 nm are assigned to $\{\bar{1}100\}$, ($\bar{1}\bar{1}2\bar{2}$), and $\{\bar{1}10\bar{1}\}$ facet-QW emissions, respectively. To quantify decay components, the time dependence was fitted by the single or double exponential equations written by

$$I(t) = A \exp(-t/\tau), \quad (3.5)$$

$$I(t) = A_{\text{fast}} \exp(-t/\tau_{\text{fast}}) + A_{\text{slow}} \exp(-t/\tau_{\text{slow}}), \quad (3.6)$$

where $I(t)$ is the PL intensity at a time of t , and τ is the lifetime. The subscripts represent the fast and slow components. The estimated lifetimes were indicated in Fig. 3.15(b). The decay curves of the ($\bar{1}\bar{1}2\bar{2}$) and $\{\bar{1}10\bar{1}\}$ facet QWs are composed of fast and slow decays, though their origins have yet to be clarified. However, it is noteworthy that ~ 3 nm thick QWs on the conventional (0001) plane exhibit 66 ns at 475 nm [36]. Thus, all the decay lifetimes of the developed 3D QWs are much faster. This strongly suggests much higher radiative recombination probabilities, which are the reciprocal of

the lifetimes, due to the weakened internal electric fields. These discussions also hold for TRPL under the selective excitation condition (370 nm): The estimated fast and slow recombination lifetimes for the $(\bar{1}\bar{1}2\bar{2})$ and $\{\bar{1}10\bar{1}\}$ facet QWs are $\tau_{\text{fast}} = 178$ ps and $\tau_{\text{slow}} = 931$ ps, and $\tau_{\text{fast}} = 281$ ps and $\tau_{\text{slow}} = 1.33$ ns, respectively. (The emission at 390 nm could not be well resolved from the 370 nm laser.)

Let us note carrier lifetimes measured at RT. Under the assumption of IQE value = 100% at LT, radiative (τ_{rad}) and nonradiative (τ_{nonrad}) lifetimes at RT can be extracted by dividing the carrier lifetime (τ_{PL}) with the IQE value at RT. Example temperature dependences of τ_{rad} and τ_{nonrad} calculated by this manner can be found elsewhere [1, 34, 37]. Therefore, τ_{PL} at RT is drastically affected by τ_{nonrad} unless the IQE at RT takes quite high values close to 100%. In other words, $\tau_{\text{PL}}(\text{RT})$ gives us insight into nonradiative processes, in contrast to that $\tau_{\text{PL}}(\text{LT})$ is a good indicator of radiative processes. Additionally, the IQE ($\eta_{\text{int}}(T)$) of emission can be written in terms of $\tau_{\text{rad}}(T)$ and $\tau_{\text{nonrad}}(T)$, where

$$\eta_{\text{int}}(T) = \frac{\tau_{\text{nonrad}}(T)}{\tau_{\text{rad}}(T) + \tau_{\text{nonrad}}(T)}. \quad (3.7)$$

This equation apparently suggests that longer $\tau_{\text{nonrad}}(\text{RT})$ is essentially required for higher $\eta_{\text{int}}(\text{RT})$, and that is indicated by longer $\tau_{\text{PL}}(\text{RT})$. A number of papers reported $\tau_{\text{PL}}(\text{RT})$ of (0001) plane InGaN QWs in the order of a few ns or more [34, 37]. On the other hand, it was found that the developed 3D QWs exhibit shorter $\tau_{\text{PL}}(\text{RT})$ below ~ 350 ps from the $(\bar{1}\bar{1}2\bar{2})$ and $\{\bar{1}10\bar{1}\}$ facet QWs, suggesting the intensive nonradiative processes compared to (0001) plane. It is, however, noted that some studies have addressed $\tau_{\text{PL}}(\text{RT})$ below ~ 1 ns of semipolar InGaN QWs on $(11\bar{2}2)$ or $\{20\bar{2}1\}$ GaN substrates [28, 33, 34]. Therefore, the shorter $\tau_{\text{PL}}(\text{RT})$ from the developed 3D QWs may not be attributed to the degradation of material quality due to the 3D faceted growth. Instead, faster nonradiative processes commonly observed in the current semipolar QWs are an important issue for the future development of semipolar-based light emitters.

3.7 Pastel Color Syntheses

Polar-plane-free faceted 3D InGaN QWs on $(\bar{1}\bar{1}2\bar{2})$ were found to exhibit radiative polychromatic emission. However, the present emission spectrum covers very limited color components within the entire visible region. Figure 3.16(a) the PL spectrum and the colors of the visible light spectrum, suggesting a lack of green or yellow emission

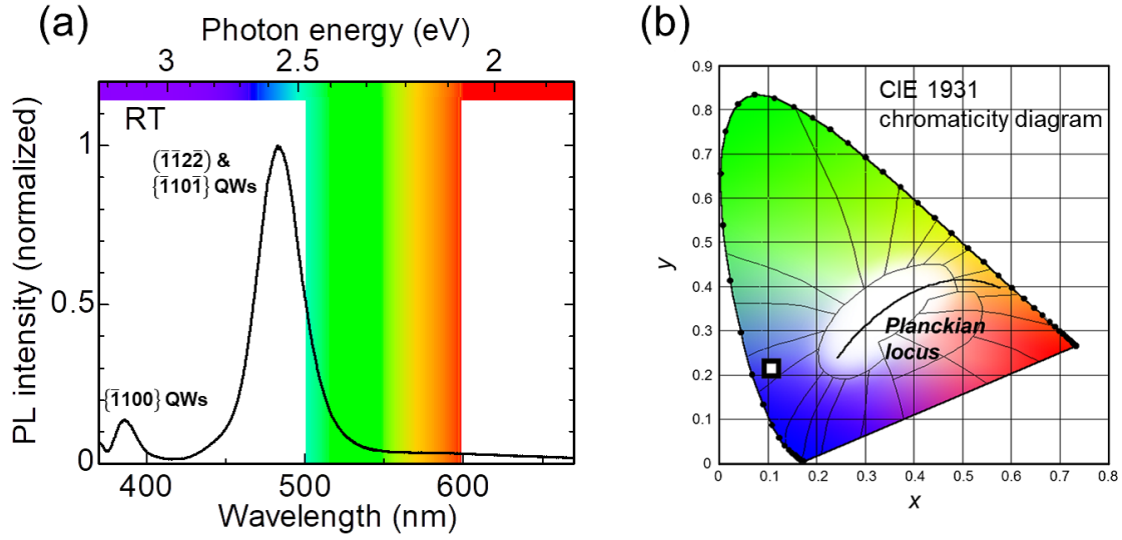


Figure 3.16: (a) PL spectrum of the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ and (b) CIE chromaticity diagram, where the open square indicates the emission from the sample.

components. In terms of the Commission Internationale de l'Éclairage (CIE) 1931 chromaticity diagram, the emission was at (0.108, 0.215), which are plotted as the open square in Fig. 3.16(b). To obtain tailored visible color spectra, it is indispensable to explore a way of covering the longer wavelength region, *i.e.*, higher In composition region. However, as reviewed in chapter 1, there are several challenges to obtain the efficient longer wavelength emissions from the In-rich InGaN systems. In this section, we investigate fundamental growth conditions to expand the emission component of the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ to longer wavelength region (green $\sim$ yellow). The facet QWs are designed, taking into account the wavefunction overlap and critical layer thickness. As a result, we demonstrate pastel green and white color syntheses from the well-designed 3D QWs on $(\bar{1}\bar{1}2\bar{2})$.

3.7.1 Structural design

Before detailed QW structural design, emission color component of each facet should be considered. The present facet QW structure is shown in Fig. 3.17. Toward longer wavelength emissions, the top $(\bar{1}\bar{1}2\bar{2})$ and inclined $\{110\bar{1}\}$ facets can be responsible for green $\sim$ yellow colors, whereas the side $\{1101\}$ facet can emit violet $\sim$ blue colors. The In compositions and well widths on each facet will be designed based on this assignment.

Semipolar QWs can improve the radiative recombination probability by the en-

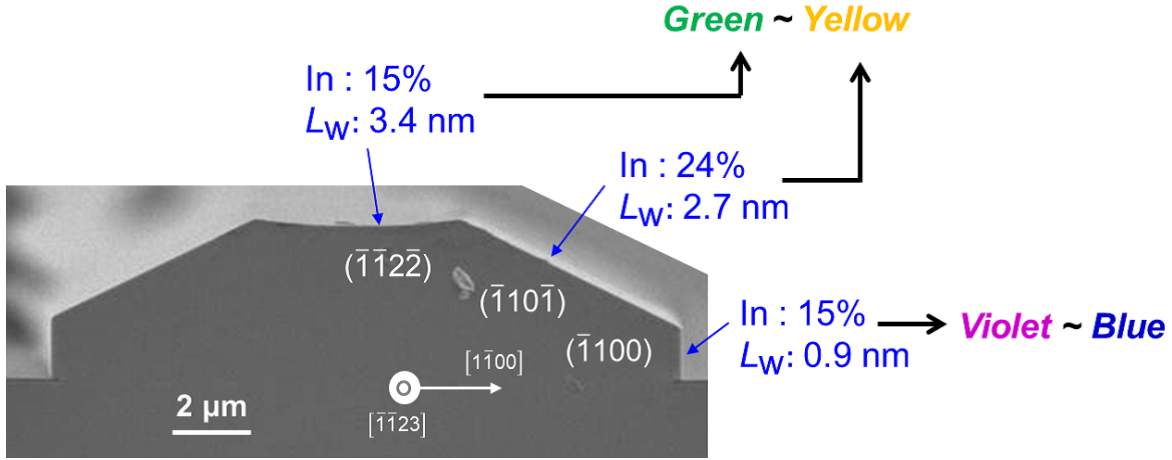


Figure 3.17: Emission color assignment for each facet of the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ toward longer wavelength emissions

hanced square of the overlap integral between the electron and hole wavefunctions, compared to the conventional (0001) polar QWs. However, in some cases, the presence of the small piezoelectric severely decreases the square overlap integral. Thus, we must carefully design the QW structural parameters of In compositions and well widths. Figure 3.18 displays the calculated transition energy at RT and the square overlap integral of InGaN/GaN QW on $\{11\bar{2}2\}$ semipolar plane as functions of the InGaN well width. The InGaN well layer is assumed to be coherently strained on the GaN layer. It should be noted that internal electric fields of $\{1\bar{1}01\}$ InGaN QWs are close to $\{11\bar{2}2\}$, and thus, the same QW structural design provides almost the same transition energy. For comparison, the calculated results of (0001) polar plane are also indicated. Regarding the transition energy, $\{11\bar{2}2\}$ (and $\{1\bar{1}01\}$) semipolar QW requires In compositions of $30 \sim 40\%$ to emit green $\sim$ yellow colors. It is noteworthy that those In compositions are higher than those of (0001) polar QW because the reduced internal electric fields of the semipolar QWs suppress the quantum-confined Stark effect (QCSE). The favorable well width can be determined from the square overlap integral. As shown in the bottom of Fig. 3.18, the overlap integral becomes smaller as the wider well width even in the semipolar QW. Smaller well width (~ 2 nm) is required to keep the higher overlap integrals.

As another viewpoint, critical layer thickness (CLT) of InGaN on GaN underlayer should also be considered. This is because, misfit dislocations (MDs) induced by lattice relaxation may work as nonradiative recombination centers and prevent high-performance light emission. Figure 3.19 shows (a) a schematic structure for CLT calcu-

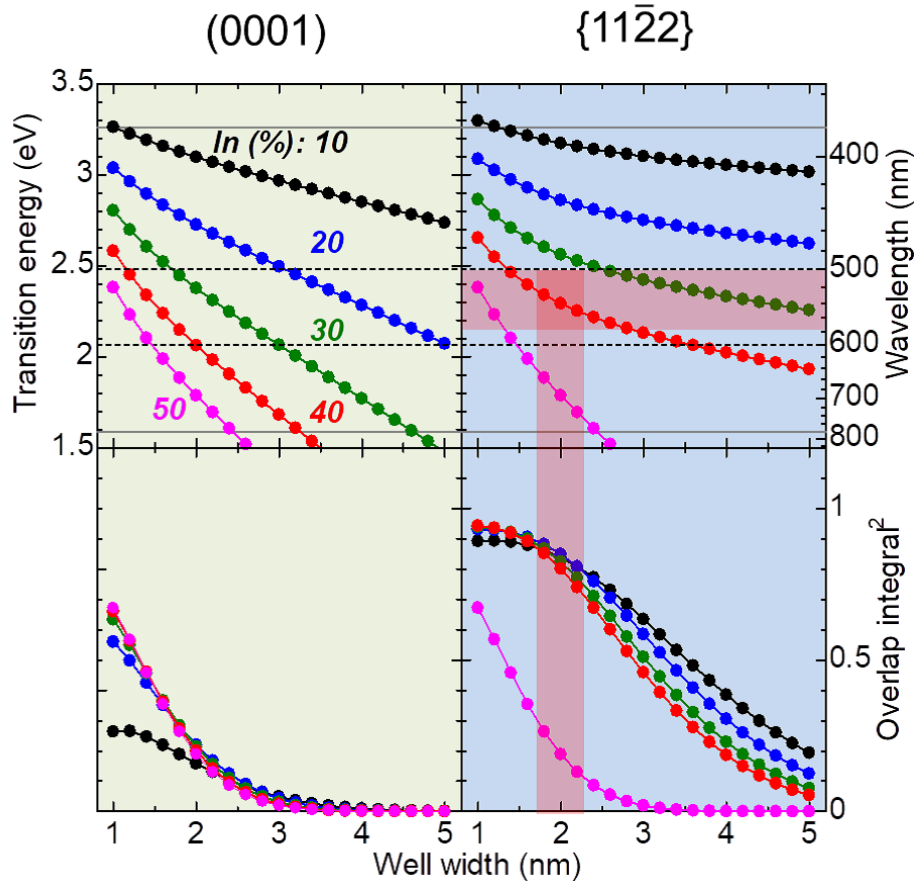


Figure 3.18: Transition energies and wavefunction overlaps of InGaN QWs on (0001) and $\{11\bar{2}2\}$ planes as a function of well width.

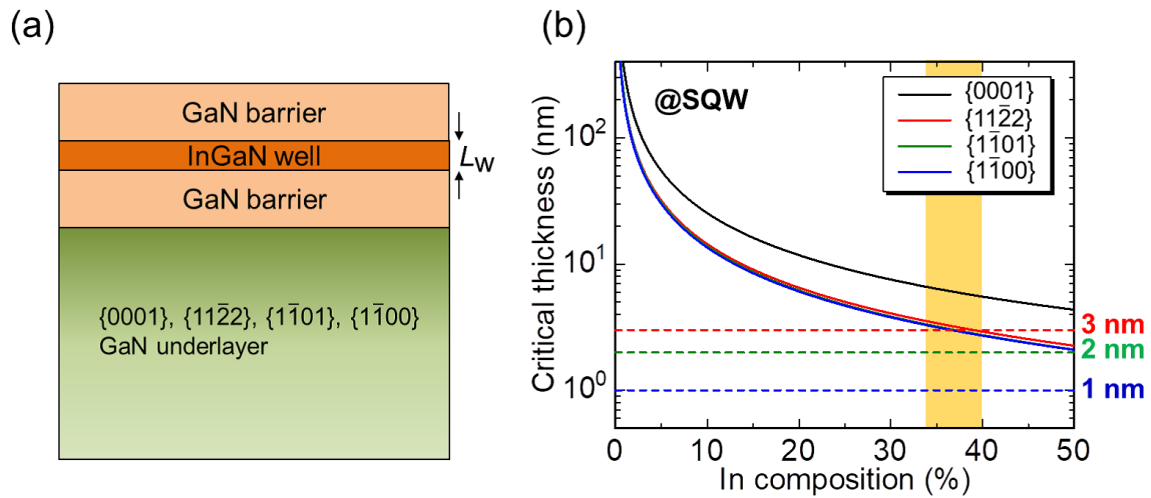


Figure 3.19: Critical layer thickness for InGaN single layers grown on various orientations. (a) A schematic illustration of InGaN QW on GaN underlayer for the calculations and (b) the results for $\{0001\}$, $\{11\bar{2}2\}$, $\{1\bar{1}01\}$ and $\{1\bar{1}00\}$ orientations.

lations and (b) the calculated results as a function of In composition. The calculations were performed based on the extended Fischer model proposed by Nishinaka *et al.* [38]. As a result, it is found that thinner well width (L_w) of around 2 nm is required to avoid the lattice relaxation in the In compositions of 30 ~ 40%. Here, we note that the thinner CLT of semipolar $\{11\bar{2}2\}$ than that of polar (0001) is one of the critical issues for semipolar-based highly efficient longer wavelength emitters because that restricts the degree of freedom for the device structure.

Let us move to the growth condition to attain the above structure design. The inter-facet well width distribution is chiefly dominated by gas phase diffusion, and lower growth temperatures aiming at higher In compositions would slightly suppress the inter-facet adatom migration. Therefore, the inter-facet well width distribution under the lower growth temperature is supposed to be almost the same as Fig. 3.17. Namely, the thickness design of ~2 nm on planar surface may provide the inter-facet thickness distribution of 2.3 nm on the top ($\bar{1}\bar{1}2\bar{2}$), 1.8 nm on the inclined ($\bar{1}10\bar{1}$), and 0.6 nm on the side ($\bar{1}100$). From these considerations, InGaN well widths were designed to be ~2 nm by reducing the growth time, and the growth temperatures were decreased to promote higher In incorporation.

3.7.2 Optical characterizations

Figure 3.20 shows (a) PL spectra from the fabricated 3D QWs at RT and (b) CIE chromaticity diagram. L_w designated in Fig. 3.20(a) is a designed InGaN well thickness on ($\bar{1}\bar{1}2\bar{2}$) planar surface. As a reference, so far discussed 3D QW is also indicated by blue line or plot. The indicated temperatures correspond to the growth temperatures of the InGaN QWs. It is found that the emission spectra from the 3D QWs are shifted to longer wavelength region by decreasing the QW growth temperatures: the emission peaks of the longer wavelength components are prolonged from ~480 nm to ~510 nm and ~570 nm, and those of the corresponding shorter wavelength components move from ~390 nm to ~410 nm and ~440 nm. Due to the additive color mixture, the apparent colors were pastel green and white, as indicated in Figs. 3.20(c) and 3.20(d). The white color is fortunately well approximated the chromaticity of blackbody radiators with a color temperature of ~4900 K, as shown in the CIE diagram.

The inferior emission characteristics caused by inadequate QW design were also observed. Figure 3.21 compares the macroscopic and spatially resolved emission properties of the 3D QWs on ($\bar{1}\bar{1}2\bar{2}$) with SQW (L_w :~2 nm) and five-period QW (L_w :~3 nm). The 5QW with the wider L_w shows the degraded emission intensity of the longer

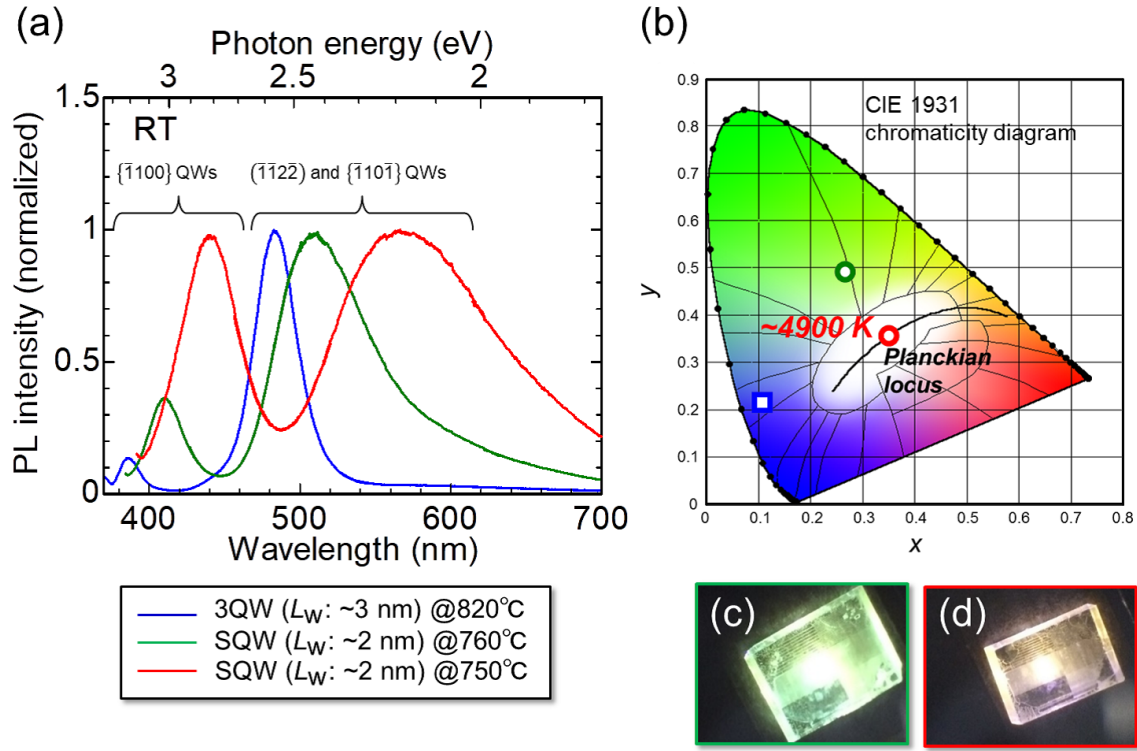


Figure 3.20: (a) PL spectra from the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ aiming at longer wavelength region and (b) CIE chromaticity diagram for the spectra. The plots in (b) are indicated by the same colors as the spectra in (a). The example photographs in (c) and (b) are the emission from the SQW grown at 760 °C and 750 °C, respectively. The white color corresponds to a color temperature of ~4900 K.

wavelength component, as shown in Fig. 3.21(a), suggesting the poor material quality derived from the lattice relaxation. The spatially resolved CL measurements reveal that the SQW with the thinner L_w emits the green emission component from the whole region of the top and inclined facets, whereas the 5QW with the wider L_w shows the emission from the limited area near the side facet.

Finally, internal quantum efficiencies (IQEs) of the 3D QWs on (0001) and $(\bar{1}\bar{1}2\bar{2})$ with similar emission spectra are compared. In order to access the IQEs, temperature dependence of PL was measured. The PL system was almost the same as Fig. 3.3, but the temperature was controlled using a closed-cycle cryostat. The excitation light source was a He-Cd (325 nm) laser with an excitation density of 31.8 W/cm². When nonradiative recombination processes are negligible at LT, the ratio of the PL intensities at RT and LT approximates the IQE at RT. Although this assumption is debatable, the PL intensity ratios are estimated to gain some insights into the IQE of the 3D QWs.

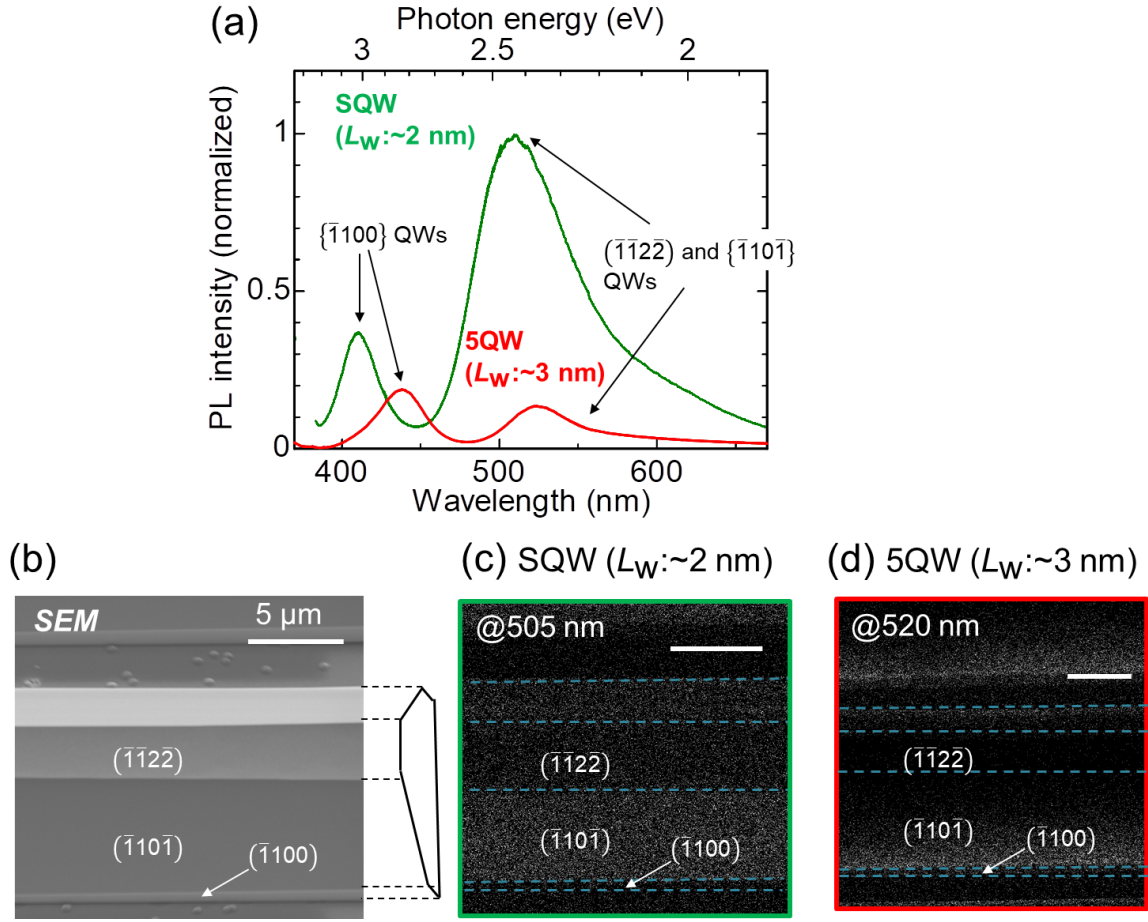


Figure 3.21: Optical property comparison between the 3D QWs on $(\bar{1}\bar{1}22)$ with the SQW ($L_w \sim 2$ nm) and 5QW ($L_w \sim 3$ nm) structures. (a) RT-PL spectra from them. (b) SEM bird's eye view of the 3D QWs and monochromatic CL mapping images taken at (c) 505 nm for the SQW ($L_w \sim 2$ nm) and (d) 520 nm for the 5QW ($L_w \sim 3$ nm). The scale bars represent 5 μm .

Figure 3.22 compares the temperature dependences of (a, b) the emission spectra and (c, d) integrated PL intensities of the 3D QWs on (0001) and $(\bar{1}\bar{1}22)$ emitting pastel green colors at RT. The 3D QWs on (0001) were grown on (0001) GaN templates with the SiO_2 mask geometry of $(w, m) = (15 \mu\text{m}, 5 \mu\text{m})$, and the 3D structure consists of the top (0001) and inclined $(11\bar{2}2)$ facets. Here, w and m are the window and mask widths. The InGaN well widths on the top (0001) and inclined $(11\bar{2}2)$ facet QWs are around 4 nm and 2 nm, respectively. For the 3D QWs on (0001) [Figs. 3.22(a) and 3.22(c)], the estimated IQE of each facet is 9.8% for the (0001) QWs and 34.2% for the $\{11\bar{2}2\}$ QWs. The higher IQE value of the violet emission than the green emission is the well-known tendency for InGaN systems, and the reduced internal electric field of

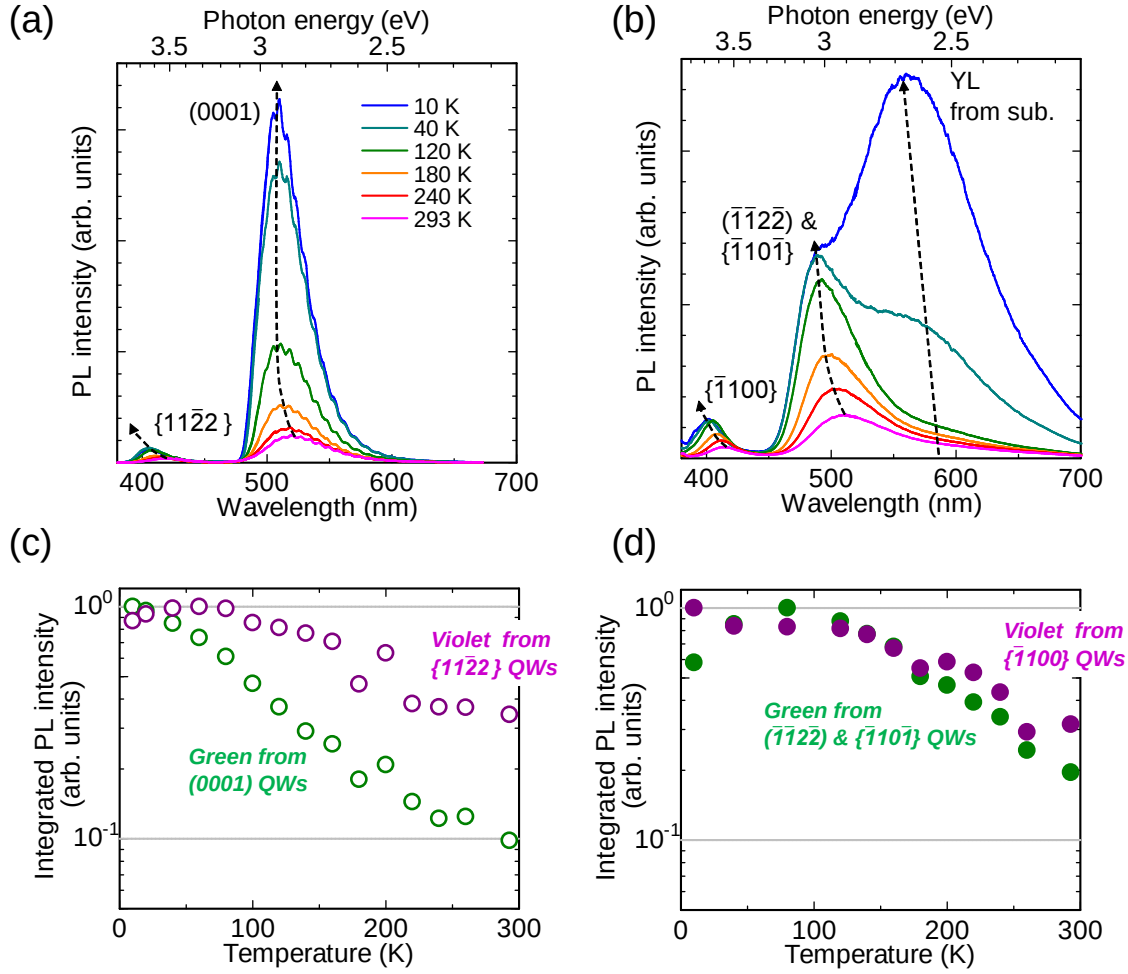


Figure 3.22: Temperature-dependent PL spectra and integrated intensity of each emission component from the 3D QWs on (a, c) (0001) and (b, d) ($\bar{1}\bar{1}2\bar{2}$).

the $\{11\bar{2}2\}$ semipolar plane may partially contribute the improvement. On the other hand, the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ exhibit the IQEs of 19.6% for the $(\bar{1}\bar{1}2\bar{2})$ and $\{\bar{1}10\bar{1}\}$ QWs and 31.5% for the $\{\bar{1}100\}$ QWs. The yellow luminescence (YL) was originated from the Si-doped GaN substrate and decomposed into Gaussian components. It is noteworthy that the IQE of the green emission from the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ is higher than that of those on (0001). One of the reasons could be attributed to the enhanced radiative recombination probability due to the reduced internal electric field of the $(\bar{1}\bar{1}2\bar{2})$ and $\{\bar{1}10\bar{1}\}$ facet QWs.

Several papers reported IQE values of the 3D faceted InGaN QWs, which ranged from 3% to 62% [1, 7, 25, 39–42]. It should be noted that IQE values severely depend on the excitation carrier densities, so the following values are just for the reference. Nishizuka *et al.* evaluated the 3D QWs on (0001) similar with this study and obtained the IQE values of $\sim 10\%$ for the (0001) facet QWs and $\sim 40\%$ for the $(11\bar{2}2)$ facet QWs [1], which is consistent with our results. Schimpke *et al.* characterized the microrod LED structures, and the reported IQE was close to 60% [41]. Very recently, Nami *et al.* reported the highest IQE value of 62% among the reported values from the core-shell nanowire LED structure. Compared with these reports, the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ show fairly large emission efficiencies, though further investigations are necessary toward the higher emission efficiencies.

3.8 Summary

InGaN QWs overgrown on the polar-plane-free 3D GaN structures on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ were investigated to explore 3D InGaN QWs exhibiting radiative polychromatic emission properties. Macroscopic PL and microscopic CL measurements revealed monochromatic and polychromatic emissions from the 3D QWs on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$, respectively. Additionally, the PL spectra indicated superior QW structural property of the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$. Local QW structural characterizations proved the inter-facet distribution of In compositions and InGaN thickness of the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$, inducing the facet-dependent polychromatic emission. The 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ and conventional (0001) planes were compared, and it was found that the inter-facet variation of effective InN thickness is quite different between them. This observation is interpreted as an influence of the crystallographic orientation dependence of In incorporation efficiency. The inclined $(\bar{1}10\bar{1})$ in the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ has significantly higher In incorporation efficiency, resulting in the suppression of surface In adatom migration. The weakened In adatom diffusion on the inclined $(\bar{1}10\bar{1})$ is also suggested from both the diffusion

equation analyses and the simplified model calculation. TRPL measurements were performed for the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ and demonstrated that all the facet QWs exhibit faster radiative lifetimes compared to (0001) polar plane. These results indicate that the polar-plane-free 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ show, indeed, semipolar and nonpolar carrier dynamics without degradation caused by the 3D growth.

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Chapter 4

Doping of Polar-Plane-Free Faceted 3D Structures

4.1 Introduction

We have demonstrated radiative polychromatic emissions using polar-plane-free faceted InGaN QWs grown on $(\bar{1}\bar{1}2\bar{2})$ semipolar GaN substrates in chapter 3. However, the results obtained under photoexcitation do not necessarily imply successful LED fabrication. Two primal issues still remain toward the LED demonstration: one is pn-junction formation on each facet, and the other is device processes on highly 3D structures. The study in this chapter is directed to investigate p-type doping conditions of the developed polar-plane-free structures.

Prior to the p-GaN growth of the polar-plane-free 3D structures, we have to establish optimal doping conditions of $(\bar{1}\bar{1}2\bar{2})$ planar LEDs. The reason is that there are several previous reports on $(11\bar{2}2)$ LEDs [1–5], whereas those on $(\bar{1}\bar{1}2\bar{2})$ LEDs are in lack (an amber LED in [1]). In addition, the $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ planes have distinct impurity incorporation behaviors. For example, the impurity incorporation of various crystallographic orientations was compared by Cruz *et al.*, and they found that $(\bar{1}\bar{1}2\bar{2})$ plane exhibits doping profile broadening of Mg and Si impurities for p-type and n-type dopants, respectively [6]. Nishinaka also observed similar doping profile broadening of Mg in $(\bar{1}\bar{1}2\bar{2})$ plane [7]. Furthermore, unintentional impurity incorporation is also different between $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ [6, 7]. These results indicate that the optimal growth conditions of $(\bar{1}\bar{1}2\bar{2})$ are different from those of $(11\bar{2}2)$ LEDs. From this background, we investigate optimal doping conditions of $(\bar{1}\bar{1}2\bar{2})$ LEDs, compared with $(11\bar{2}2)$ LEDs. We demonstrate that $(\bar{1}\bar{1}2\bar{2})$ LEDs with bright electroluminescence (EL) can be obtained

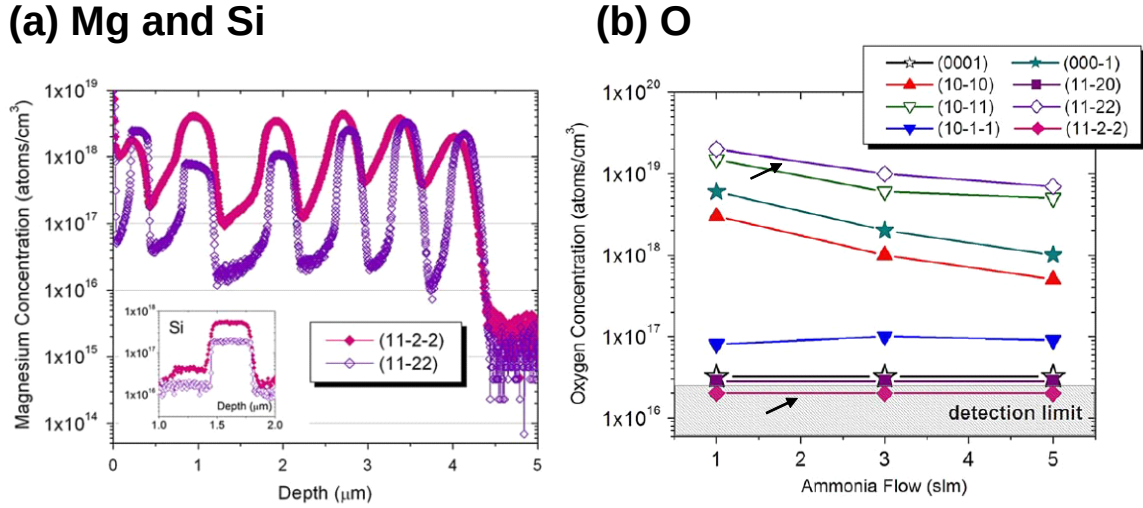


Figure 4.1: (a) Mg and Si SIMS profiles of $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ homoepitaxially-grown GaN, and (b) NH_3 flow dependence of O concentrations in undoped GaN films grown on various crystallographic orientations cited from Ref. [6].

by optimizing II/III ratios and growth pressures.

After establishing doping conditions of $(\bar{1}\bar{1}2\bar{2})$ planar LEDs, the growth condition is employed to the growth of polar-plane-free 3D LED structures on $(\bar{1}\bar{1}2\bar{2})$. Here, one of the primal issues is pn-junction formation on all facets of the 3D structures. Although there are several reports of p-GaN growth on 3D structures grown on (0001) GaN templates [8–11], the processes that govern the inter-facet variations of p-GaN thickness or Mg concentration have yet to be understood, comprehensively. In this study, we demonstrate the pn-junction formation of the 3D LED structures on $(\bar{1}\bar{1}2\bar{2})$ and discuss the inter-facet p-GaN thickness and incorporated Mg concentration.

4.2 Doping Conditions of $(\bar{1}\bar{1}2\bar{2})$ Planar LEDs

Crystallographic orientations strongly affect impurity incorporation behaviors, resulting in quite a different diode performance among them grown under the same growth condition, likely due to the surface bond configurations. Figure 4.1(a) shows secondary ion mass spectroscopy (SIMS) depth profiles of Mg and Si for simultaneously grown $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ oriented layers investigated by Cruz *et al* [6]. The samples were deposited alternating Mg-doped (Si-doped) GaN layers grown at different growth conditions with undoped GaN layers. The Mg and Si doping profiles were sharp at layer interfaces for the $(11\bar{2}2)$ orientations. On the other hand, $(11\bar{2}2)$ plane shows higher

concentrations of O, as shown in Fig. 4.1(b). These observations suggest that optimal growth conditions for $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ LEDs would be quite different.

In this section, we firstly investigate LED device properties of $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ LEDs grown under the same growth condition, in order to establish $(\bar{1}\bar{1}2\bar{2})$ LED growth conditions. As expected, distinct device performance is observed: the $(\bar{1}\bar{1}2\bar{2})$ LEDs show smaller turn-on voltages and series resistances, but the electroluminescence (EL) intensities are quite weaker than that of the $(11\bar{2}2)$ LEDs. SIMS analyses reveal that the poor doping profiles of $(\bar{1}\bar{1}2\bar{2})$ LEDs degrade the EL intensities but, at the same time, suggest a way of improving doping profiles. Finally, we achieve bright EL from $(\bar{1}\bar{1}2\bar{2})$ LEDs with proper impurity profiles by adjusting p-type doping conditions.

4.2.1 Experiments

The samples were grown on $(11\bar{2}2)$ or $(\bar{1}\bar{1}2\bar{2})$ n-GaN substrates with the carrier density of $\sim 10^{18} \text{ cm}^{-3}$. The source precursors were trimethylgallium (TMGa), triethylgallium (TEGa), NH_3 , monosilane (SiH_4) for Si doping, and bis-cyclopentadienyl magnesium (Cp_2Mg) for Mg doping. Figure 4.2 shows (a) a LED structure design and (b) temperature and pressure profiles examined in this study. From the viewpoint of fundamental characterization, the LED structure consists simply of Si-doped n-GaN (3 μm), InGa N (3 nm)/Ga N (20 nm) single QW (SQW), Mg-doped p-GaN (300 nm), and heavily Mg-doped p^+ -Ga N (20 nm). Unlike chapter 3, the III-precursor for the InGa N QW layer was changed to TEGa due to the MOVPE system's maintenance during this study. The TEGa flow rate was preliminarily adjusted, yielding InGa N well and Ga N barrier growth rates similar to those of previous TMGa. The growth pressure was varied between 300 Torr and 650 Torr, according to the doping profile optimization. The reason will be described below.

For the p-GaN layer, where is the subject in this study, TEGa and NH_3 were used as Ga and N sources, respectively, and the carrier gas was H_2 . The relatively lower growth temperature ($\sim 930^\circ\text{C}$) was employed to suppress In desorption from the InGa N well layer and/or the degradation during p-GaN growth [12]. The growth pressures and flow ratios of Mg and Ga sources (II/III ratios) were varied for doping profile optimization, and several combinations of pressure and II/III ratio presented in Fig. 4.2(b) were examined. Here, II/III ratios were changed by adjusting Mg flow rates under constant Ga flow rates. For sequence A (Seq. A), the combination of pressures P (Torr) and II/III ratios were $(P, \text{II/III}) = (350, 2.0\text{e-}3)$ or $(400, 5.0\text{e-}3)$. For Seq. B, P or II/III ratios were independently increased from $(400, 5.0\text{e-}3)$ in Seq. A, and the investigated

combinations are (450, 5.0e-3), (500, 5.0e-3), (550, 5.0e-3), and (400, 7.0e-3). Finally, II/III ratios were decreased, and p-GaN layers were grown under the conditions of (550, 1.4e-3), (550, 2.0e-3), (650, 1.4e-3), and (650, 2.0e-3) in Seq. C. It should be noted that, in Seq. C, the pressures were increased during the temperature transition from n-GaN to InGaN QW. After MOVPE growth, the samples were annealed at 650 °C in air for 10 min to activate to Mg acceptors in p-GaN layers. It is noted that the activation anneal condition was preliminarily optimized for (11 $\bar{2}2$) p-GaN and was not optimized for ($\bar{1}\bar{1}2\bar{2}$) p-GaN.

Vertical LED designs were employed utilizing n-type conductivity of {11 $\bar{2}2$ } GaN substrates. This is a little off topic in this study, but let us note that vertical LED design is expected to possess superior current spreading than lateral LED design; spatial mismatch between the p-contact and n-contact in conventional lateral design lead to severe current crowding in some cases [13–15]. Indeed, Cao *et al.* experimentally demonstrated a reduced series resistance of a vertical LED compared to a lateral LED [16]. Electrodes were formed by standard UV photolithography and resistance heating vacuum evaporation. Firstly, AlN pastes remaining on the sample backsides were removed by rapping technique. Then, the samples were dipped in HCl : H₂O (1 : 1) at 100 °C for 10 min to remove the oxides on the surfaces. After the cleaning, Ni (5 nm)/Au (10 nm) p-contact and Ti (50 nm)/Au (200 nm) p-pad electrodes were formed by the vacuum evaporation using lift-off photolithography processes. Here, Ni/Au p-contact layers were annealed at 650 °C in air for 10 min. Finally, Ti (30 nm)/Al (100 nm) n-contact and Ti (50 nm)/Al (200 nm) n-pad electrodes were deposited from the backsides, followed by annealing at 450 °C in air for 10 min to form ohmic contact to n-GaN. The chip size was 350×350 μm^2 , and the LEDs were on wafer without packages.

4.2.2 Comparison between simultaneously grown (11 $\bar{2}2$) and ($\bar{1}\bar{1}2\bar{2}$) LEDs

First of all, (11 $\bar{2}2$) and ($\bar{1}\bar{1}2\bar{2}$) LEDs with the growth conditions of Seq. A are compared to extract a clue for doping conditions of ($\bar{1}\bar{1}2\bar{2}$) LEDs. Figure 4.3 shows (a) current-voltage (I - V) characteristics and (b) EL spectra acquired from the (11 $\bar{2}2$) and ($\bar{1}\bar{1}2\bar{2}$) LEDs. The growth condition for the p-GaN layers is (P , II/III) = (400, 5.0e-3). The experiments were performed under direct current (DC) operation at room temperature (RT). In Fig. 4.3(a), both the (11 $\bar{2}2$) and ($\bar{1}\bar{1}2\bar{2}$) LEDs show clear rectifying properties, suggesting pn-junctions were successfully formed. In addition, the ($\bar{1}\bar{1}2\bar{2}$) LED has smaller turn-on voltage and series resistance than those of the (11 $\bar{2}2$) LED. The EL

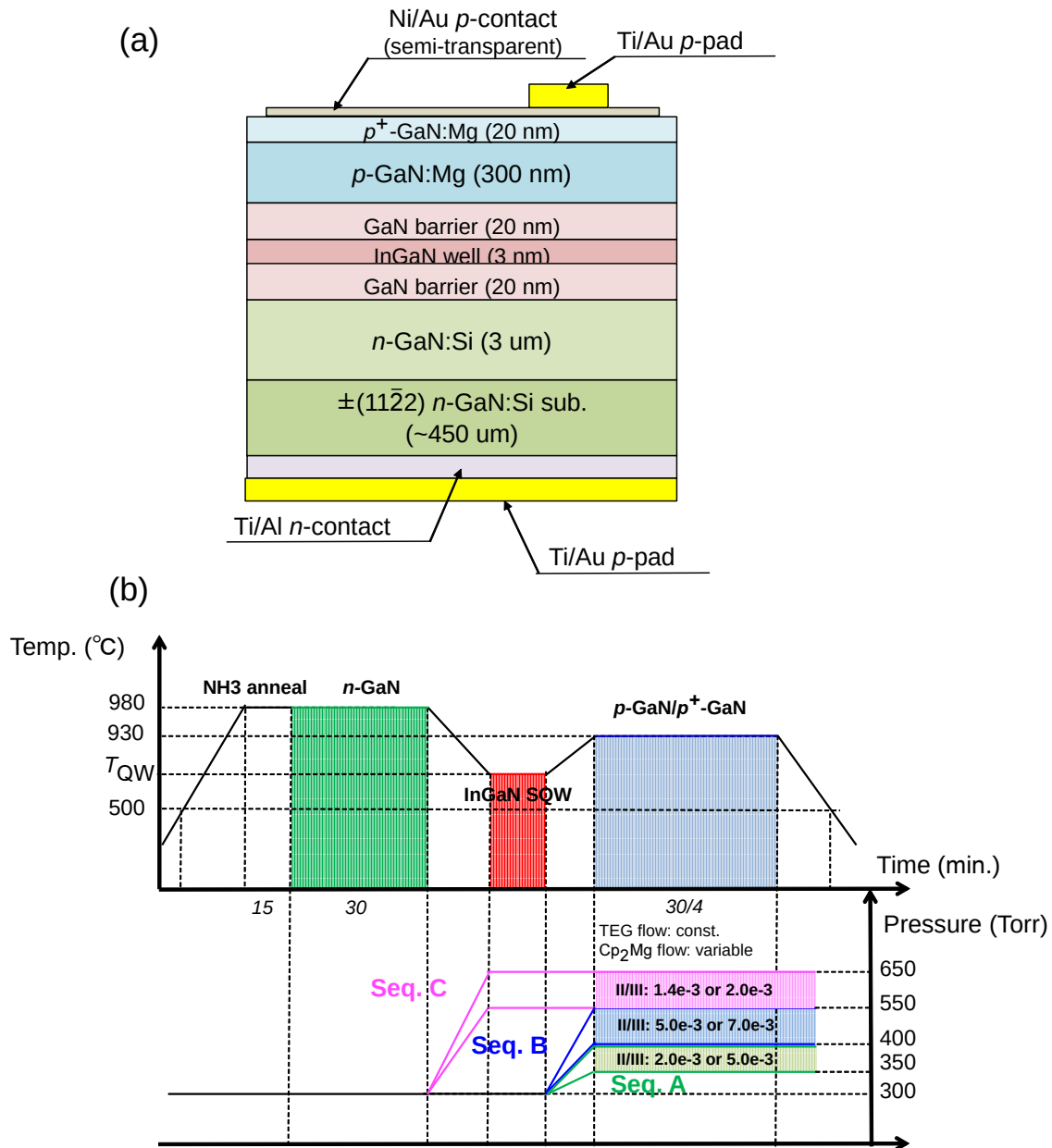


Figure 4.2: (a) Schematic cross-section of the $\pm(11\bar{2}\bar{2})$ planar LED structure, and (b) temperature and pressure sequences used in this study. Using the same temperature profiles, growth pressure sequences and II/III ratios are varied between the samples. Seq. A is used to compare the device performance of the $(11\bar{2}\bar{2})$ and $(\bar{1}\bar{1}\bar{2}\bar{2})$ LEDs. In Seq. B, growth pressure or II/III ratio are independently increased up to 550 Torr or 7.0e-3, respectively, from (400, 5.0e-3) used in Seq. A. In Seq. C, higher pressures (550 Torr or 650 Torr) with smaller II/III ratios (1.4e-3 or 2.0e-3) are used.

intensity from the $(\bar{1}\bar{1}2\bar{2})$ LED is, however, quite weaker than that from the $(11\bar{2}2)$ LED, as shown in Fig. 4.3(b). The inset photographs in Fig. 4.3(b) apparently shows the bright EL from the $(11\bar{2}2)$ LED, whereas nearly invisible EL of the $(\bar{1}\bar{1}2\bar{2})$ LED. Similar trends were observed for the comparison between the $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ LEDs with $(P, \text{II/III}) = (350, 2.0\text{e-}3)$.

To clarify the origins of the quite different device properties between the $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ LEDs, the depth profiles of intentional impurities, Si and Mg, and unintentional impurities, O and C, were analyzed by SIMS, using Cs^+ as the primary ion at an acceleration voltage of 5 kV. Depth profiles of In were also obtained to identify the position of InGa_N QWs. All SIMS measurements throughout this study were performed by the Foundation for Promotion of Material Science and Technology of Japan (MST). Here, it should be noted that O in MOVPE GaN acts as a shallow donor with substituting N sites [17]. On the other hand, C seems to work as compensators in both n-GaN and p-GaN via substituting N sites: C impurities in n-GaN and p-GaN form acceptor-like and donor-like states, respectively [18, 19]. Figure 4.4(a) and 4.4(b) shows SIMS depth profiles of the $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ LEDs with the p-GaN growth condition of $(400, 5.0\text{e-}3)$, respectively. For the $(11\bar{2}2)$ LEDs [Fig. 4.4(a)], sharp Si and Mg doping profiles and quite low C concentrations were obtained. The Si and Mg concentrations, $[\text{Si}]$ and $[\text{Mg}]$, are $\sim 1.7 \times 10^{18} \text{ cm}^{-3}$ in n-GaN and $\sim 8 \times 10^{19} \text{ cm}^{-3}$ in p-GaN, respectively. The C concentration $[\text{C}]$ is found to be $\sim 7 \times 10^{15} \text{ cm}^{-3}$ in n-GaN and $\sim 2 \times 10^{16} \text{ cm}^{-3}$ in p-GaN. Although the higher O concentration $[\text{O}]$ in the order of 10^{19} cm^{-3} is observed along the entire region, $[\text{Mg}]$ in p-GaN $\simeq 8 \times 10^{19} / \text{cm}^{-3}$ is larger than $[\text{O}] \simeq 2 \times 10^{19} \text{ cm}^{-3}$. Therefore, the bright EL from the $(11\bar{2}2)$ LED in Fig. 4.3(b) can be attributed to the effective doping profile. The factors that induce the higher turn-on voltage and series resistance is still under investigation, but one of the possible reasons is Fermi-level pinning at the p-GaN/p-contact interface [20].

On the other hand, the $(\bar{1}\bar{1}2\bar{2})$ LED exhibits poor doping property, as shown in Fig. 4.4(b). $[\text{Si}]$ in n-GaN and $[\text{Mg}]$ in p-GaN are $\sim 6 \times 10^{18} \text{ cm}^{-3}$ and $\sim 8 \times 10^{19} \text{ cm}^{-3}$, respectively, which are comparable to those of the $(11\bar{2}2)$ LED [Fig. 4.4(a)], but the profile broadenings are observed at around the InGa_N QW. This may degrade the EL intensity due to the following reasons. One is that Si and/or Mg impurities diffused into QWs create nonradiative recombination centers, resulting in low internal quantum efficiency (IQE) of the InGa_N QWs. The other is that the InGa_N QW locates outside the depletion layer due to the delay of the Mg incorporation, consequently disturbing efficient recombination of electrons from n-GaN and holes from p-GaN in the InGa_N QW. In addition to the inferior Si and Mg profiles, O and C profiles also suggest the poor

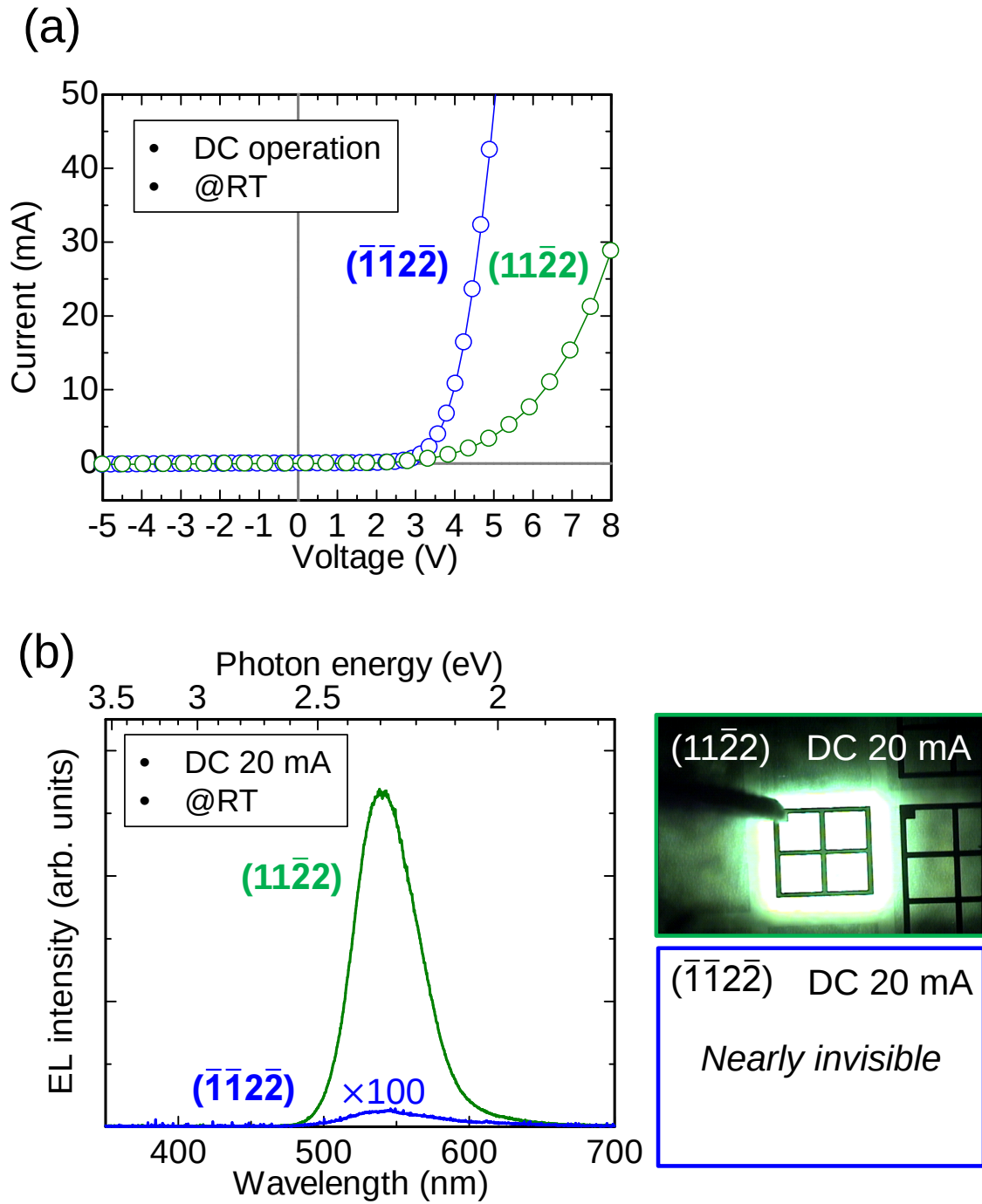


Figure 4.3: (a) I - V characteristics and (b) EL spectra of $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ LEDs. The growth conditions of the p-GaN layers are II/III (5.0×10^{-3}) and growth pressure (400 Torr).

doping property. [C] in QW is much higher than that in the $(11\bar{2}2)$ LED. Furthermore, both [O] and [C] drastically increase in the p-GaN layer, which would lead to ineffective p-type doping. Therefore, the weak EL intensity from the $(\bar{1}\bar{1}2\bar{2})$ LED in Fig. 4.3(b) is considered to be caused by the Si and Mg profile broadening and/or the higher [O] and [C] in the epi-layers.

Although the doping profile of the other p-GaN growth condition, (350, 2.0e-3), also has essentially the same trend, the profile comparisons between them indicate a way of improving doping conditions. Figure 4.5 compares SIMS depth profiles of (a) Mg and (b) O and C grown under the two different growth conditions. For Mg profiles [Fig. 4.5(a)], Mg profile broadening is slightly improved under the higher growth pressure and II/III ratio. Meanwhile, the higher pressure and II/III ratio increase [O] and [C] in the p-GaN layers, as shown in Fig. 4.5(b). This observation means that pressures and II/III ratios strongly affect the doping profiles, though the lower [O] and [C] with (350, 2.0e-3) are still higher than a proper doping profile. The reasons for these growth condition dependences will be discussed later, but these findings suggest the possibility of obtaining proper doping profiles via controlling growth pressures and II/III ratios. In the following, we investigate LED device properties grown under various II/III ratios and pressures.

4.2.3 Exploring doping conditions for $(\bar{1}\bar{1}2\bar{2})$ LEDs

Based on the growth condition of (400, 5.0e-3), growth pressure and II/III ratio were independently increased, as shown in Seq. B of Fig. 4.2(b), and the device properties were characterized. As a representative result, the device performance of (550, 5.0e-3) is shown in Fig. 4.6. Compared to the $(\bar{1}\bar{1}2\bar{2})$ LED with (400, 5.0e-3) [Fig. 4.3(a)], the similar I - V curve was observed. On the other hand, the EL intensity was slightly improved, and the emission became more visible as an indicated photograph in Fig. 4.3(b). This result indicates that higher growth pressure would be promising for improving doping profiles. However, the EL spectrum has multi-peaks from near band-edge emission at 366 nm, donor-acceptor pair (DAP) emission at around 410 nm, and broad QW emission at around 490 nm, and the DAP emission intensity is larger than the QW emission. In addition, we observed the creamy surface near the sample edge. It is well known that excessively-doped GaN:Mg layers result in surface morphology deteriorations [21]. Thus, these results suggest that higher growth pressures with lower II/III ratios, *i.e.*, lower Mg flow rates in this study, would be effective, and thus, Seq. C in Fig. 4.2(b) was employed.

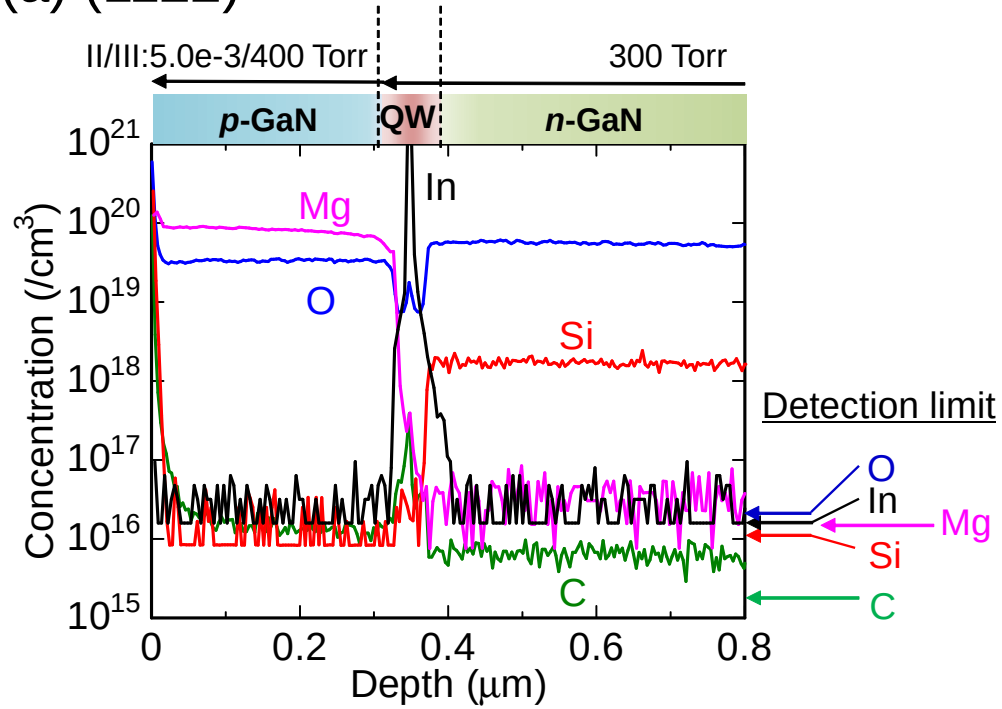
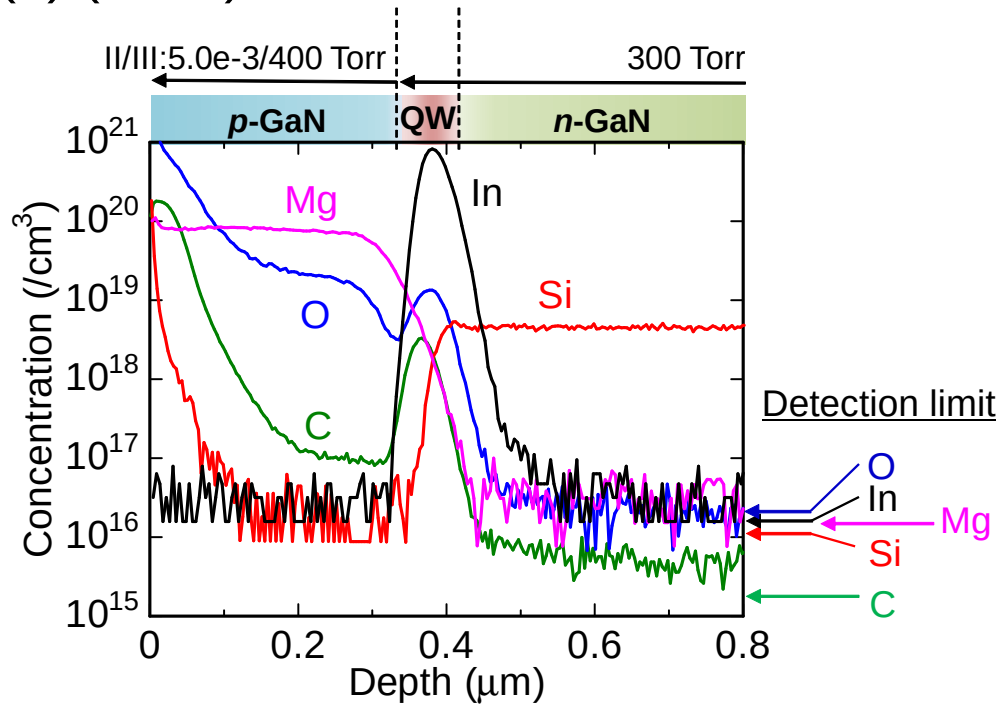
(a) $(11\bar{2}2)$ (b) $(\bar{1}\bar{1}2\bar{2})$ 

Figure 4.4: SIMS depth profiles of (a) $(11\bar{2}2)$ and (b) $(\bar{1}\bar{1}2\bar{2})$ LEDs. The p-GaN growth condition is growth pressure of 400 Torr and II/II ratio of 5.0e-3.

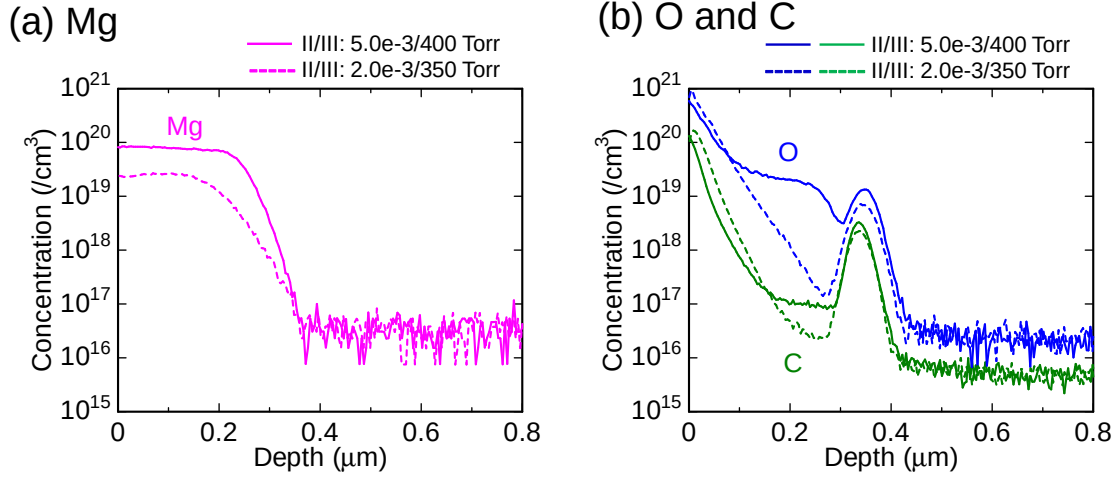


Figure 4.5: SIMS depth profile comparisons between the $(\bar{1}\bar{1}2\bar{2})$ LEDs grown under the two different p-GaN growth conditions of Seq. A. (a) Intentional impurity, Mg, and (b) unintentional impurities, O and C.

Here, it is noted that when the II/III ratio was increased to $7.0\text{e-}3$ with 400 Torr, the $(\bar{1}\bar{1}2\bar{2})$ LED also showed a slightly improved EL intensity. However, a multi-peak EL spectrum similar to Fig. 4.6(b) was observed, and the sample edge seemed to be a creamy surface. This observation also motivated us to the growth condition of Seq. C.

Figure 4.7 summarizes (a) representative I - V characteristics and (b) EL spectra grown under the different p-GaN growth conditions of Seq. A, Seq. B, and Seq. C. Each growth condition is (400, $5.0\text{e-}3$) for Seq. A, (550, $5.0\text{e-}3$) for Seq. B, and (550, $1.4\text{e-}3$) for Seq. C. The device properties of Seq. A and Seq. B have been already shown in Fig. 4.3 and Fig. 4.6, respectively. It should be noted that, in Seq. C, the pressure was increased after the growth of n-GaN layers. From Fig. 4.7(a), Seq. C has a higher turn-on voltage and series resistance compared with Seq. A and Seq. B. However, it is noteworthy that the EL intensity from the LED with Seq. C is the largest among them. Additionally, the EL spectrum is almost single peaks at ~ 480 nm from the QW, without near band-edge emission or DAP emission. This indicates the efficient carrier injections into the InGaN QWs. The broad spectrum at around 600 nm is photoluminescence (PL) originating from the photoexcited substrate via blue EL. Moreover, the mirror-like surface was observed in the entire region of the sample. The example photograph shown in Fig. 4.7(c) looks bright blue luminescence. The magnified image in Fig. 4.7(d) shows purplish-white due to the saturation of a detection camera. These superior EL characteristics suggest an excellent doping profile of the $(\bar{1}\bar{1}2\bar{2})$ LED with Seq. C. The growth conditions examined in this study and the

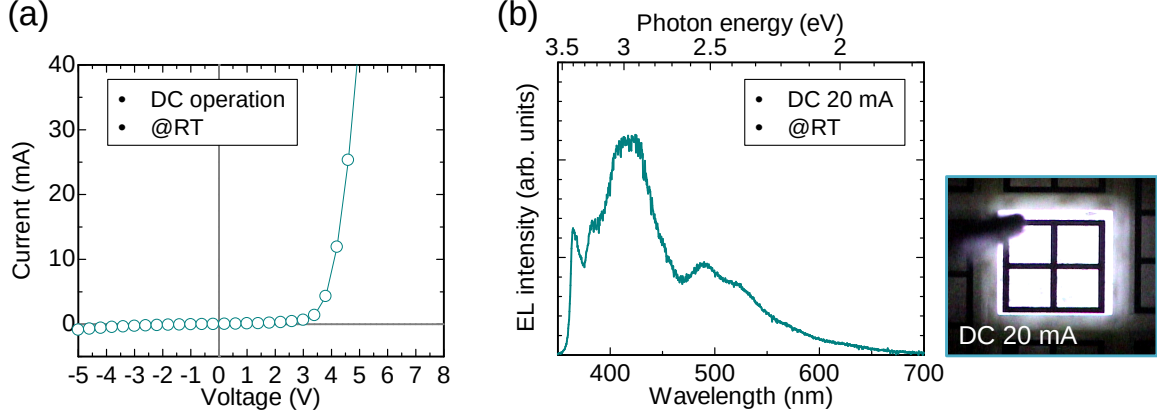


Figure 4.6: (a) I - V characteristic and (b) EL spectrum of a $(\bar{1}\bar{1}2\bar{2})$ LED. The growth condition of the p-GaN layer is II/III ratio of $5.0\text{e-}3$ and growth pressure of 550 Torr.

representative photographs are summarized in Fig. 4.8.

SIMS analysis was performed to the sample with Seq. C (550, $1.4\text{e-}3$), and the result is presented in Fig. 4.9(a). In the comparison with Seq. A in Fig. 4.4(b), the Mg and Si profile broadenings are drastically suppressed, though the Mg profile still shows a slight incorporation delay. It is, however, worth noting that the Mg profile broadening of the improved $(\bar{1}\bar{1}2\bar{2})$ LED is comparable to that of a standard (0001) p-GaN grown on a (0001) GaN template fabricated in our laboratory, as shown in Fig. 4.9(b). The (0001) InGaN LED with this p-GaN growth condition provides fairly good performance with a turn-on voltage of ~ 2.8 V [22]. Meanwhile, the $(11\bar{2}2)$ plane shows very sharp Mg profiles among them. Fichtenbaum *et al* also observed sharper Mg profiles of N-polar (000 $\bar{1}$) GaN:Mg layers than that of Ga-polar (0001) GaN:Mg layers [23]. Since the discussion on surface atom arrangements in Sec. 2.5.3 suggests the adatom kinetics of the $(11\bar{2}2)$ plane similar to the (000 $\bar{1}$) plane, this trend is likely due to the orientation dependence of O incorporation. In addition to the Mg and Si profile improvements of the optimized $(\bar{1}\bar{1}2\bar{2})$ LED, [O] and [C] in the p-GaN layer are also drastically suppressed to each detection limit of $\sim 1 \times 10^{16} \text{ cm}^{-3}$ and $\sim 1.5 \times 10^{16} \text{ cm}^{-3}$, respectively. The quite low [C] value is significant because, to the best of our knowledge, that would be the lowest level among the reported values [23, 24]. From the above results, the bright EL in Fig. 4.7(b) can be attributed to the significantly improved doping profiles.

We note that, in this study, the growth condition of (550, $1.4\text{e-}3$) is tentatively considered to be the best among the four kinds of doping conditions in Seq. C [Fig. 4.8]. In terms of the EL characteristics, the other growth conditions show slightly

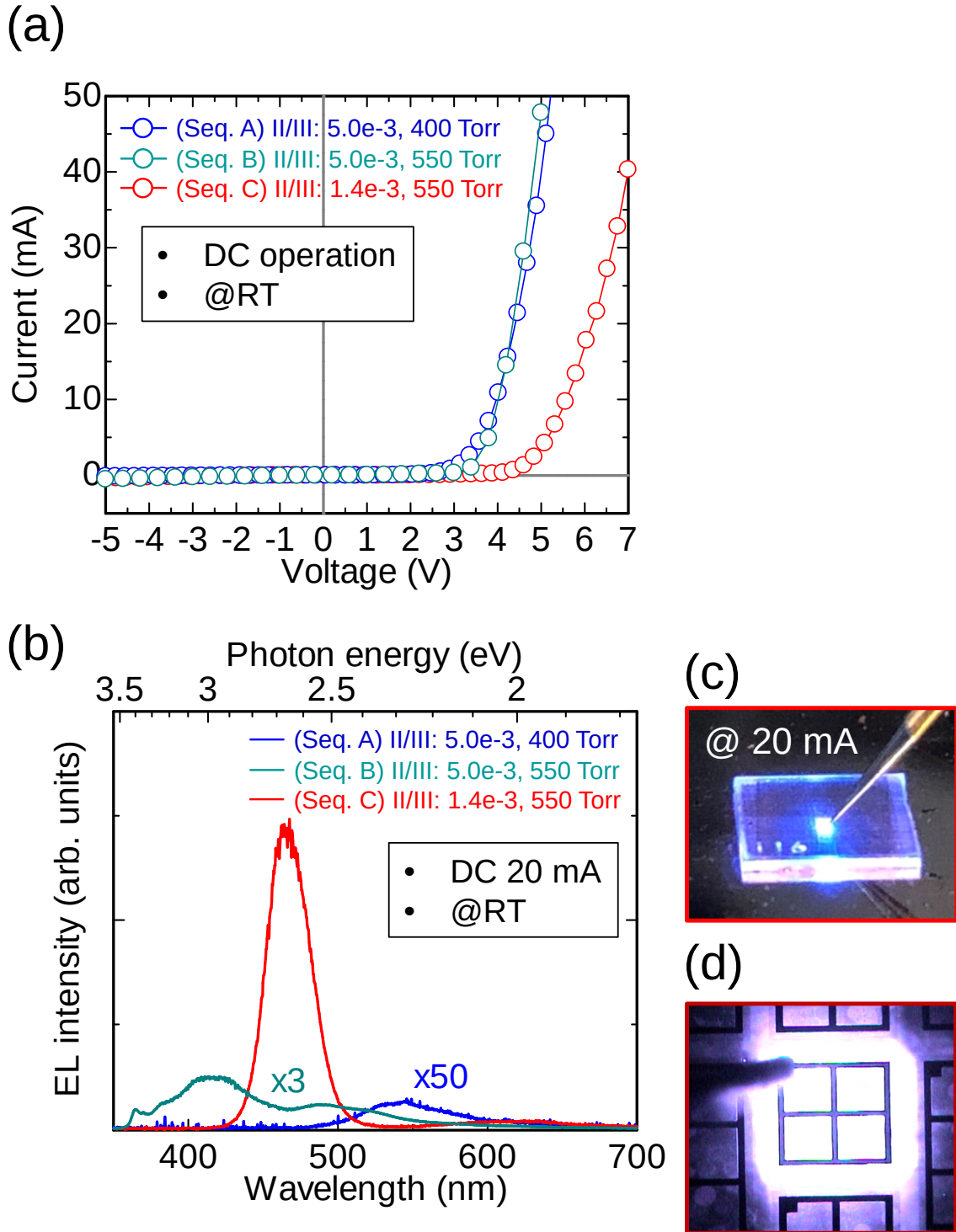


Figure 4.7: (a) I - V characteristics and (b) EL spectra of $(\bar{1}\bar{1}2\bar{2})$ LEDs grown under the different growth conditions. (c) Example photograph of the emission from the $(\bar{1}\bar{1}2\bar{2})$ LED with Seq. C at DC 20 mA, and (d) the magnified image.

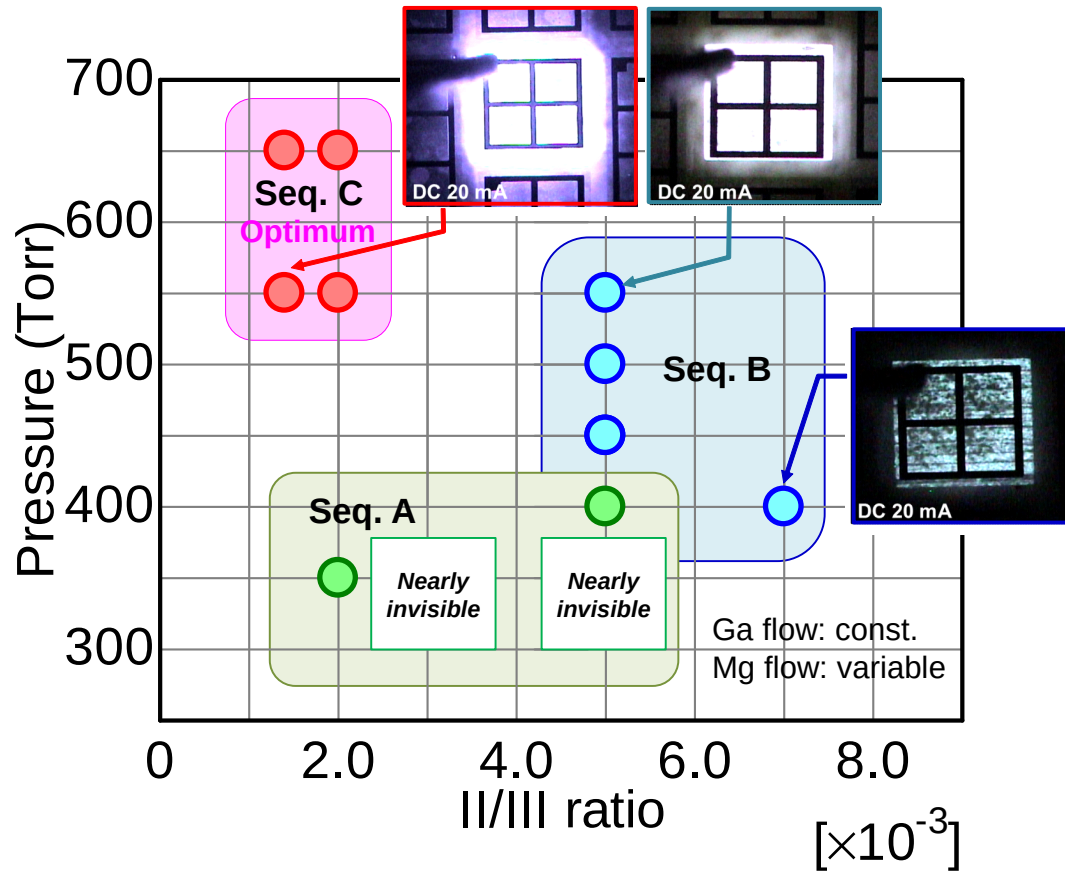


Figure 4.8: $(\bar{1}\bar{1}2\bar{2})$ LED samples varying II/III ratios and growth pressures in p-GaN growth. It should be noted that growth pressures were increased from QW layers in Seq. C.

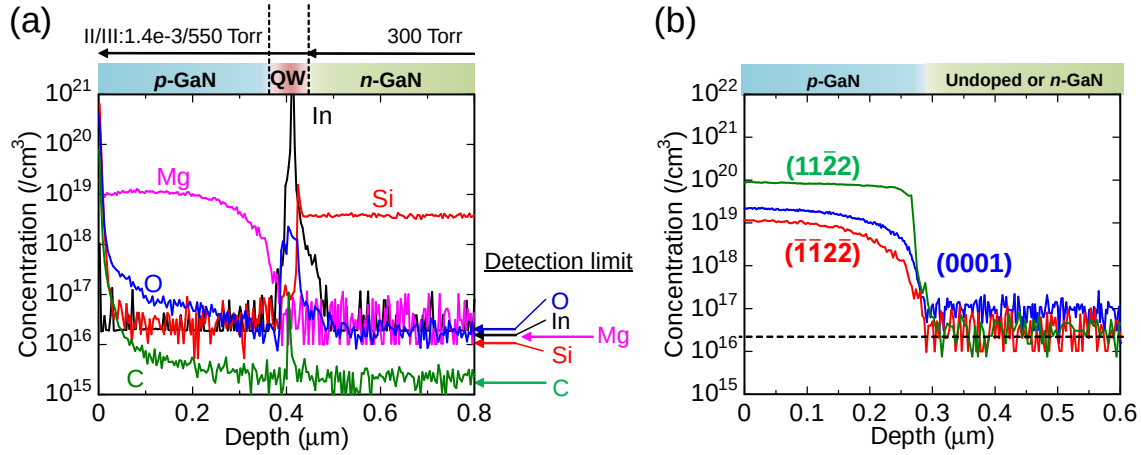


Figure 4.9: SIMS depth profile of (a) an optimized $(\bar{1}\bar{1}2\bar{2})$ LED. The II/III ratio and growth pressure are $1.4\text{e-}3$ and 550 Torr, respectively. (b) Mg profiles of the $(\bar{1}\bar{1}2\bar{2})$ LED in (a), the $(11\bar{2}2)$ LED in Fig. 4.4(a), and a p-GaN layer on a (0001) GaN template. The growth condition for the (0001) p-GaN is II/III ratio of $1.2\text{e-}3$ and pressure of 300 Torr, which is a standard condition in our laboratory.

larger DAP emission or smaller QW emission intensities than $(550, 1.4\text{e-}3)$. However, the difference is not very significant, and thus, it is difficult to draw a conclusion about the best growth condition in Seq. C. A future experimental study is expected.

4.2.4 Impurity incorporation behavior of $(\bar{1}\bar{1}2\bar{2})$ plane

We finally achieved bright EL from the $(\bar{1}\bar{1}2\bar{2})$ LED via doping profile optimization, through adjusting growth pressures and II/III ratios. The improved doping profile shown in Fig. 4.9(a) is obtained, but it is unclear which growth parameters, pressures and II/III ratios, affect Mg and Si profile broadenings and [O] and [C] reductions. Here, let us discuss the growth condition dependences of each impurity incorporation. To make clear the growth dependences, SIMS profiles of the representative samples with growth conditions $(400, 5.0\text{e-}3)$ from Seq. A, $(550, 5.0\text{e-}3)$ from Seq. B, and $(550, 1.4\text{e-}3)$ from Seq. C are compared and summarized in Fig. 4.10. It should be noted that the pressure is increased from Seq. A (400 Torr) to Seq. B (550 Torr) with the same II/III ratios of $5.0\text{e-}3$, and the II/III ratio is decreased from Seq. B ($5.0\text{e-}3$) to Seq. C ($1.4\text{e-}3$) with the same pressures of 550 Torr. Therefore, the pressure dependence of impurity incorporation can be extracted from the comparison between the samples with Seq. A and Seq. B, and the II/III ratio dependence can be extracted from the comparison between the samples with Seq. B and Seq. C.

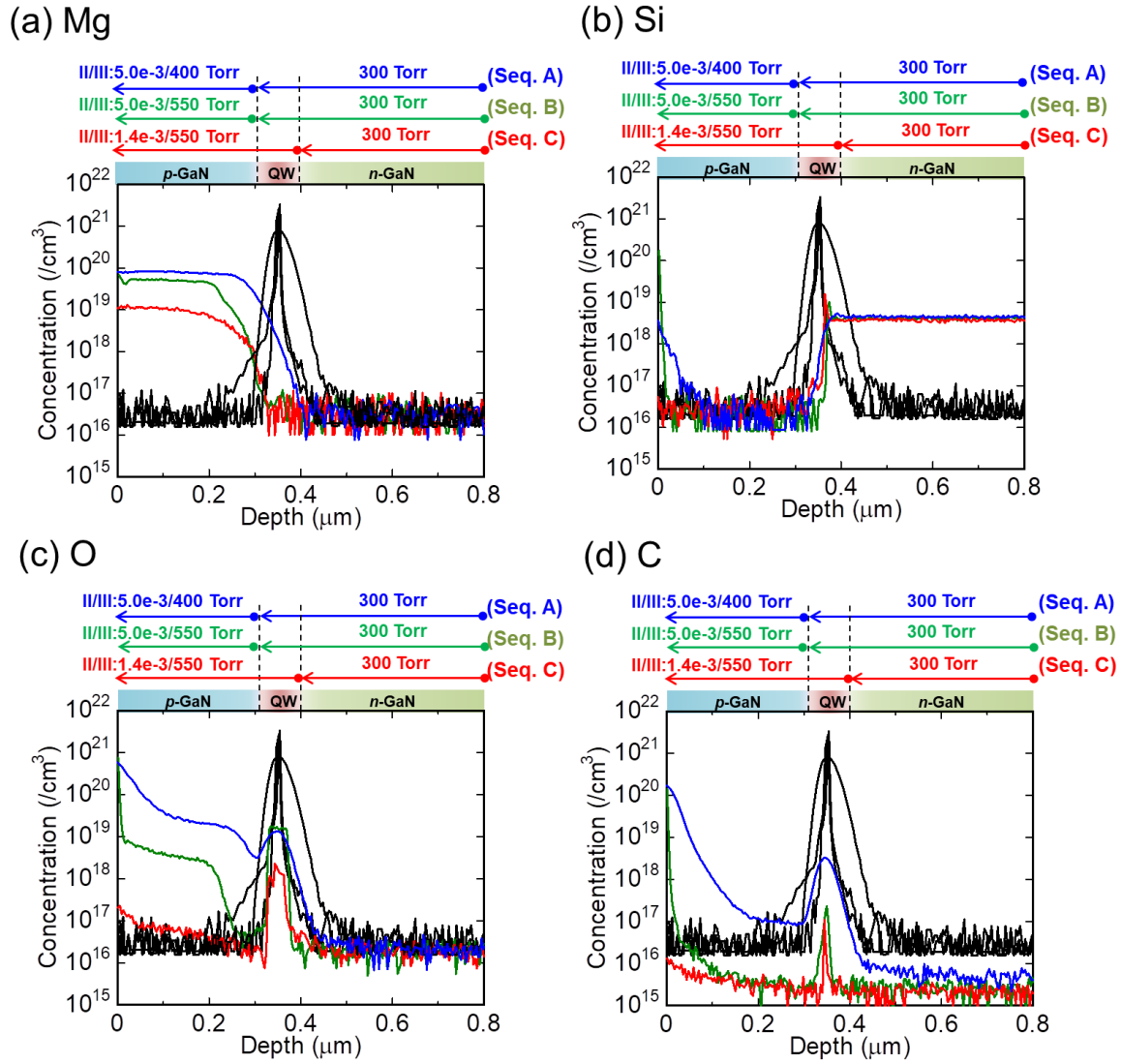


Figure 4.10: SIMS depth profiles of (a) Mg, (b) Si, (c) O, and (d) C for the $(\bar{1}\bar{1}2\bar{2})$ LEDs grown under the three different growth conditions, Seq. A, Seq. B, and Seq. C.

For Mg profiles [Fig. 4.10(a)], comparing Seq. A and Seq. B, the higher pressure of the p-GaN layer is considered to improve the profile broadening, whereas the profiles are independent of the II/III ratios from Seq. B. and Seq. C. Therefore, the EL improvement indicated in Fig. 4.7(b) can be attributed to the higher pressure. However, we also found that much higher pressure (650 Torr) delays the Mg incorporation again, indicating that there is an optimal pressure for Mg incorporation. A similar trend can be found in Si profiles [Fig. 4.10(b)], and higher pressures cause sharper Si profiles. Furthermore, the profile improvement was observed even when the pressure was increased from p-GaN layer in Seq. B, indicating Si atom diffusion during p-GaN growth under lower pressure (350~400 Torr).

For the unintentional impurity of O [Fig. 4.10(c)], both II/III ratio and growth pressure are important for the concentration reduction. Comparing Seq. A with Seq. B, [O] in QW and p-GaN layers is found to be decreased by one order of magnitude by the higher pressure. Then, from Seq. B and Seq. C, the lower II/III ratio provides even less [O] in p-GaN layers around the detection limit ($\sim 2.0 \times 10^{16} \text{ cm}^{-3}$). Similar behaviors for pressure dependence of O incorporation have already been reported on (0001) undoped GaN films [23, 25]. The proposed reduction mechanism is that higher growth pressures lead to more active nitrogen to the growth surface, and consequently, the incorporation of O substituting N sites decreases. On the other hand, to the best of our knowledge, there is no report for II/III ratio dependence of O incorporation. One possible scenario is that Cp_2Mg precursor $(\text{C}_5\text{H}_5)_2\text{Mg}$ has much amount of carbon atoms and reacts with H_2O included in NH_3 gas, forming molecules with O atoms in the gas phase, which diffuse into the growth surfaces. If this is the case, a lower II/III ratio, *i.e.*, lower Cp_2Mg flow in this study, would directly decrease [O]. For C impurity [Fig. 4.10(d)], [C] in p-GaN layer is found to be drastically decreased under higher growth pressures. Similar trend can be found in [23, 25, 26]. The reason is considered to be similar with the case of O profiles; the higher pressures promote active nitrogen to the growth front, suppressing incorporation of C substituting N sites. On the other hand, [C] in the QW layer is suppressed even under the growth condition of Seq. B, where the pressure was increased from the p-GaN layer. This is also observed in the Si profiles and the reason is under investigation.

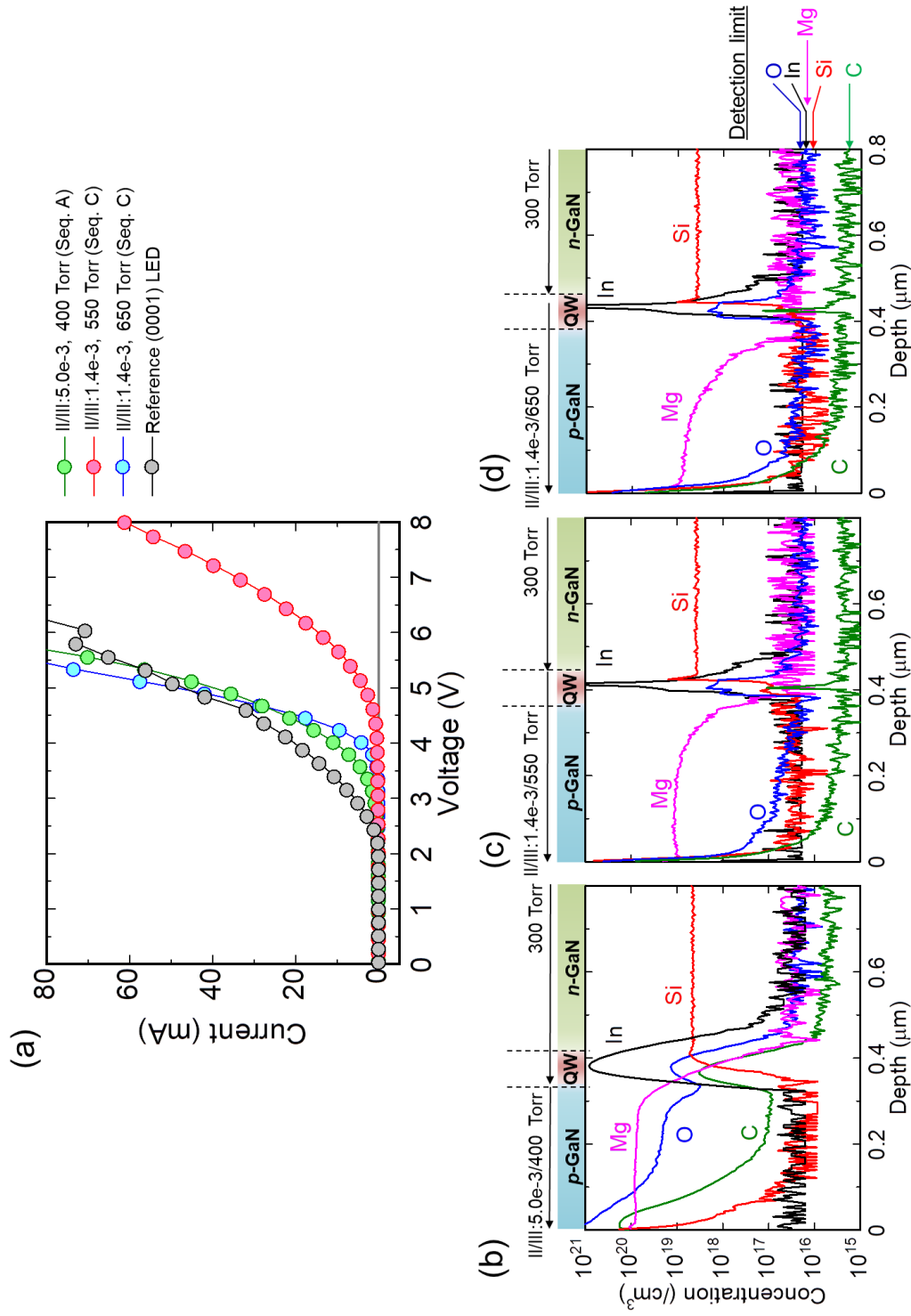


Figure 4.11: (a) I - V characteristics and (b-d) SIMS depth profiles of $(\bar{1}\bar{1}2\bar{2})$ LEDs grown under the different doping conditions. (b) (400, 5.0e-3) of Seq. A, (c) (550, 1.4e-3) of Seq. C, and (d) (650, 1.4e-3) of Seq. C. As a reference, typical I - V characteristic of a (0001) InGaN LED fabricated in our laboratory is also indicated.

4.2.5 Current-voltage characteristics under different doping conditions

We will move to the discussion on the doping condition dependence of the I - V characteristics. As already indicated in Fig. 4.7(a), different doping conditions severely vary the turn-on voltages or series resistances. Therefore, the typical I - V characteristics under different doping conditions are compared in Fig. 4.11, in an attempt to make clear the dominant factors. The samples with the doping conditions of (400, 5.0e-3) from Seq. A, (550, 1.4e-3) and (650, 1.4e-3) from Seq. C were selected for this comparison. The reason is that (400, 5.0e-3) from Seq. A and (650, 1.4e-3) from Seq. C exhibit the similar I - V characteristics, though the doping profiles are quite different. Then, (550, 1.4e-3) and (650, 1.4e-3) from Seq. C show the similar doping profiles, though the distinct I - V characteristics are observed.

Firstly, the fairly good rectifying behavior of (400, 5.0e-3) from Seq. A suggests that, in our case, the poor doping profiles in the p-GaN layer do not necessarily degrade the diode performance. The reason is under investigation, but one of the plausible factors is that the remarkably higher impurity concentrations near the p-GaN surface might suppress the Fermi level pinning; the “efficiently Mg-doped” (11 $\bar{1}2\bar{2}$) p-GaN usually shows the higher turn-on voltage due to the Fermi level pinning [20], as also shown in Fig. 4.3. This, in turns, indicates the similar Fermi level pinning in ($\bar{1}\bar{1}2\bar{2}$) p-GaN. According to this consideration, the I - V characteristic of (550, 1.4e-3) from Seq. C, showing the higher turn-on voltage and the larger series resistance, can be regarded as the behavior of the efficiently Mg-doped ($\bar{1}\bar{1}2\bar{2}$) p-GaN. The degraded diode behavior is attributed to the high barrier height derived from the pinning effect. On the other hand, the superior I - V characteristic of (650, 1.4e-3) from Seq. C seems to be inconsistent at the first time. However, looking at the outermost surface, the impurity concentrations are larger than those of (550, 1.4e-3) from Seq. C, which could also suppress the pinning effect. In addition to this, the doping profile of (650, 1.4e-3) from Seq. C presents the region with quite low impurity concentrations above the QW layer, owing to the Mg incorporation delay. Despite this resistive layer, the I - V behavior is almost the same as (400, 5.0e-3) from Seq. A. These observations suggest that in our case the p-GaN surface states which impact the barrier heights between p-contact and p-GaN, should dominantly affect the I - V behaviors.

The amounts of the barrier heights due to the Fermi level pinning are also indicated from the I - V behaviors of the p-GaN layers. Electrical properties of p-contact on p-GaN in the ($\bar{1}\bar{1}2\bar{2}$) LED structures were evaluated using the experimental configuration

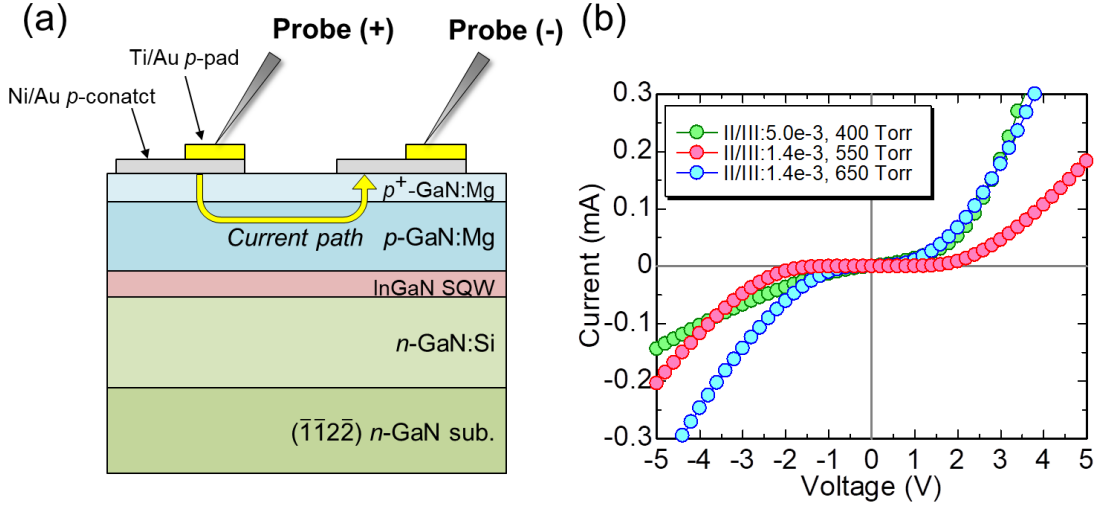


Figure 4.12: (a) Schematic of experimental setup measuring electrical properties of p-contact on p-GaN of the $(\bar{1}\bar{1}2\bar{2})$ LEDs and (b) the obtained I - V characteristics.

shown in Fig. 4.12(a). The obtained results in Fig. 4.12(b) indicates that the p-GaN grown under (550, $1.4\text{e-}3$) from Seq. C has non-ohmic contact and larger series resistance, suggesting the higher barrier height compared to the other p-GaN growth conditions. This trend is consistent with the above discussion.

4.3 Doping in 3D LED Structures on $(\bar{1}\bar{1}2\bar{2})$

In section 4.2, the growth condition of InGaN LED structures on $(\bar{1}\bar{1}2\bar{2})$ plane has been established. Therefore, let us move to the growth of 3D LED structures on $(\bar{1}\bar{1}2\bar{2})$ plane. One of the critical issues is to form pn-junction in the 3D structures entirely. On the other hand, it is also difficult to electrically evaluate the conductivity of 3D faceted structures due to the textured surface. In this study, scanning electron microscopy (SEM) is basically performed to characterize spatial dopant distributions of the 3D structures. In addition to this, another approach based on scanning probe microscopy (SPM) technique is proposed in an attempt to clarify the local electrical properties in the cross sections of the 3D structures.

4.3.1 Experiments

The 3D LED structures were grown on $(\bar{1}\bar{1}2\bar{2})$ GaN substrates. The fabrication procedures for selective area growth are essentially the same as described in Sec. 2.2. Figure

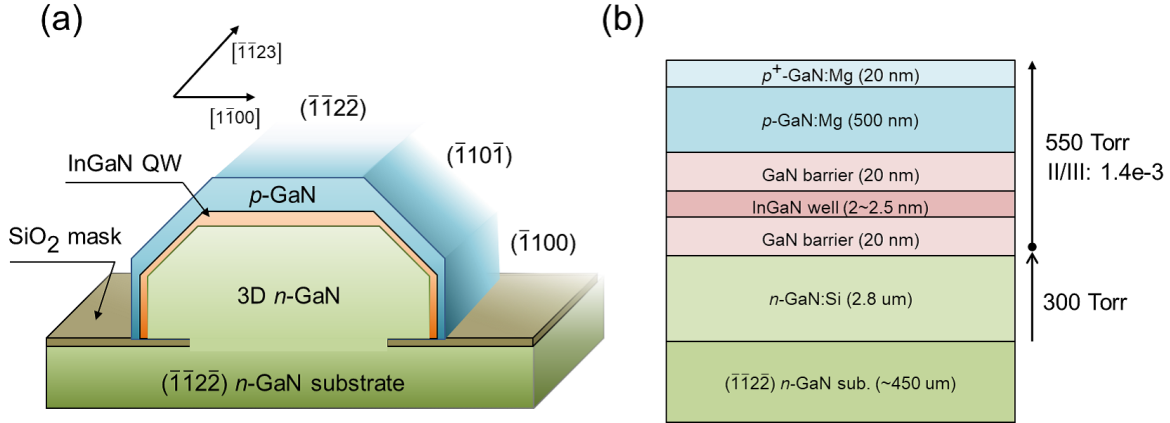


Figure 4.13: Schematics of (a) a polar-plane-free faceted 3D LED structure and (b) the thickness design on $(\bar{1}\bar{1}2\bar{2})$ planar surface.

4.13(a) shows a schematic design of a 3D LED structure. The thickness design for each epi-layer on a planar surface is depicted in Fig. 4.13(b). It is noted that the designed p-GaN thickness was increased to 500 nm from that (300 nm) of the $(\bar{1}\bar{1}2\bar{2})$ planar LED [Fig. 4.2(a)] by adjusting the growth time, aiming at sufficient p-GaN thickness (~ 100 nm) on the $(\bar{1}\bar{1}00)$ side facet. This is because, the $(\bar{1}\bar{1}00)$ facet has the slowest growth rate among all the facets in 3D GaN structures and InGaNs QWs [Figs. 2.10(f) and 3.6(g)]. The other growth parameters were the same as the Seq. C in Sec. 4.2. Firstly, Si-doped n-GaN was grown at 300 Torr, and subsequently, InGaN/GaN SQW, Mg-doped p-GaN, and heavily Mg-doped p-GaN layers were grown at 550 Torr. The II/III ratio was set to 1.4e-3. The InGaN QW was designed to emit pastel green color. (Detailed QW structural parameters can be found in Sec. 3.7.)

4.3.2 Structural characterizations

We note that undoped MOVPE GaN usually exhibits n-type conductivity with a carrier concentration of $10^{15} \sim 10^{17} \text{ cm}^{-3}$, and can easily increase the concentration up to around 10^{18} cm^{-3} via Si doping. The situation is different for p-type doping. More precise control of growth conditions is required for Mg-doped p-GaN, even on planar surfaces, as discussed in section 4.2. In addition to this doping issue, three-dimensionality is an obstacle to evaluate the conductivity because it is technically difficult to electrically confirm the p-type conductivity of GaN:Mg layer on each facet in 3D structures, by a commonly used Hall measurement for planar films. Conventional SIMS analysis, which also gives us effective information on the doping situation in a sample, is also

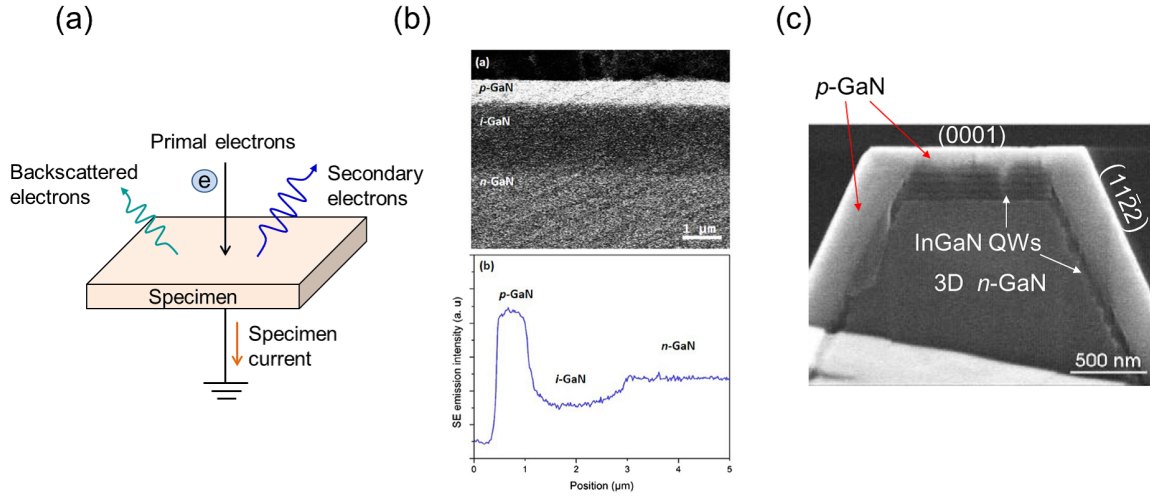


Figure 4.14: (a) Schematic diagram of emitted electrons from the specimen in SEM. (b) SE image and the SE intensity profile of the p-i-n GaN structure cited from Ref. [28]. (c) SEM photograph of a 3D LED structure cited from Ref. [29]. Assigns for each layer or facet planes are added by the author.

unsuitable due to the in-plane spatial resolution, typically $\geq 10 \mu\text{m}$. Recently, some promising characterization methods for micro- or nano-structures are emerging, such as electron beam induced current microscopy (EBIC) [27] or atom probe tomography (APT). However, these techniques require elaborate sample preparations and are very time-consuming. From these backgrounds, in this study, SEM observation is chosen for characterizing dopant distribution in the 3D LED structures on $(\bar{1}\bar{1}2\bar{2})$. In the following, the basic idea is briefly described.

SEM contrast

Secondary electrons (SE) in a SEM can provide information about spatial dopant distribution. Figure 4.14(a) shows a schematic diagram illustrating emitted electrons from the specimen when the incident electron beam (primary electrons) enters the specimen. When the primary electrons enter the specimen, SEs are produced from the emission of the valence electrons of the constituent atoms in the specimen. Since the energy of SE is very small, those generated at a deep region are quickly absorbed by the specimen, whereas those generated at the top surface of the specimen are emitted outside of the specimen. That means that SE is very sensitive to the surface, and thus, the SE is used to observe the topography of the specimen surface. Here, the SE emission in a specimen involves three steps, starting with the generation of SE inside the bulk by the incident primary electrons, their transport toward the surface, and their emission

by escaping into vacuum. These steps depend on the diffusion length of electrons in the bulk, the mean escape depth of electrons near the surface, the band bending due to surface states, and their chemical nature of the surface. These factors depend on the doping characteristics of the semiconductor and the chemical nature of the surface, resulting in doping level dependence of the SE emission.

Recently, Alugubelli *et al.* comprehensively investigated SE emission intensity profiles of p-i-n GaN structures and figured out that p-type GaN shows larger SE emission intensity, resulting in brighter SEM contrast compared to n-type GaN, as shown in Fig. 4.14(b) [28]. Similar tendency has been also reported in Si, SiC, and GaAs systems [30–33]. Alugubelli *et al.* attributed the reason to the downward and upward band bending near the surface of p-GaN and n-GaN, respectively. Downward band bending of p-GaN promotes electron transport to the surface, thus enhancing SE emission, whereas upward band bending near the surface of n-GaN prevents electron transport to the surface, thus decreasing SE emission. The brighter SEM contrast of a p-GaN layer has been empirically known so far, and Neubert *et al.* confirmed p-GaN layer overgrown on 3D GaN structures fabricated on (0001) GaN templates, as shown in Fig. 4.14(c). Our group also reported similar observation [34].

results and discussions

Figure 4.15(a) shows cross-sectional SEM images of a fabricated 3D LED structure on ($\bar{1}\bar{1}2\bar{2}$). The accelerating voltage was set to 5 kV through all experiments in this section. Mg-doped p-GaN layers with brighter SEM contrast can be observed above Si-doped 3D n-GaN structure with darker SEM contrast. Although the bright contrast cannot be well resolved at the side ($\bar{1}100$) facet, the magnified image of the corner in Fig. 4.15(b) indicates a thin p-GaN layer on the side facet. From the contrast, the p-GaN thickness on each facet is measured to be $\sim 1.3 \mu\text{m}$ on the top ($\bar{1}\bar{1}2\bar{2}$) facet, $\sim 300 \text{ nm}$ on the inclined ($\bar{1}10\bar{1}$) facet, and $\sim 100 \text{ nm}$ on the side ($\bar{1}100$) facet, which yields the relative growth rates of $v_{(\bar{1}\bar{1}2\bar{2})} : v_{(\bar{1}10\bar{1})} : v_{(\bar{1}100)} = 13 : 3 : 1$. These observations lead us to conclude that pn-junction formation on each facet is successfully achieved. However, the drastic inter-facet thickness variation may cause an obstacle to equivalent carrier injections to all facets.

Then, p-GaN growth conditions were varied in order to improve the thickness uniformity. In 3D GaN structures on (0001), some papers reported that local V/III on growth surface is considered to strongly affect 3D shapes [35, 36]. Therefore, V/III ratios were varied in the range of 5600 - 8500 with changing the NH_3 flow rate during p-GaN growth. However, the lateral growth rates were not significantly increased.

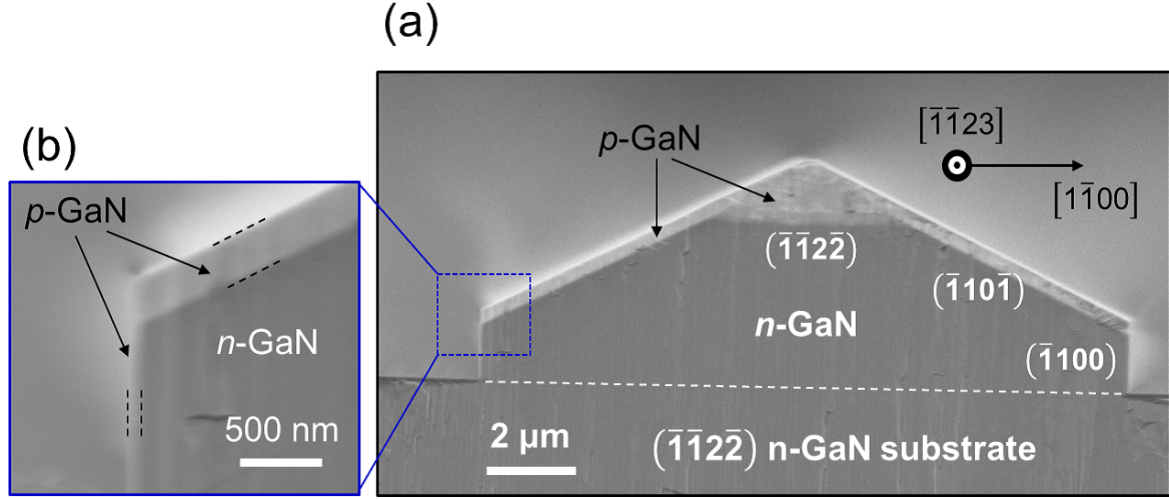


Figure 4.15: Cross-sectional SEM images of (a) a fabricated 3D LED structure on $(\bar{1}\bar{1}2\bar{2})$ and (b) the magnified image of the $(\bar{1}\bar{1}0\bar{1})$ and $(\bar{1}\bar{1}00)$ facets.

Alternatively, lower II/III ratio with decreased Mg flow rate was examined. Tanikawa *et al.* reported that hillocks observed on the surface of $(000\bar{1})$ GaN can be suppressed by Mg doping, which enhances surface adatom migration [37]. If this is also the case in this study, the p-GaN thickness distributions would be affected by the Mg doping, and lower II/III ratios may result in suppressed inter-facet adatom migration from the inclined $(\bar{1}\bar{1}0\bar{1})$ to the top $(\bar{1}\bar{1}2\bar{2})$ facets. Thus, additional 3D LED structures were grown under the II/III ratio of $1.0\text{e-}3$ by decreasing the Mg flow rates. It should be noted that the p-GaN growth time was set to 40 min in this experiment. The cross-sectional SEM is presented in Fig. 4.16. The inter-facet variation of p-GaN thickness is as follows: $\sim 1.0\text{ }\mu\text{m}$ on $(\bar{1}\bar{1}2\bar{2})$, $330\sim 400\text{ nm}$ on $(\bar{1}\bar{1}0\bar{1})$, and $\sim 90\text{ nm}$ on $(\bar{1}\bar{1}00)$, yielding the relative growth rates of $v_{(\bar{1}\bar{1}2\bar{2})} : v_{(\bar{1}\bar{1}0\bar{1})} : v_{(\bar{1}\bar{1}00)} = 11.1 : \sim 4.1 : 1$. Focusing on the growth rate relationship between the $(\bar{1}\bar{1}2\bar{2})$ and $(\bar{1}\bar{1}0\bar{1})$ facets, the relative growth rate of p-GaN on the $(\bar{1}\bar{1}2\bar{2})$ is slightly decreased compared with the previous p-GaN growth condition in Fig. 4.15. Therefore, the lower II/III ratio might suppress the inter-facet surface diffusion, but it still exhibits drastic thickness variation. We need further investigations toward uniform p-GaN thickness distributions.

On the other hand, it is interesting to note that the distinct SEM contrasts are observed between the p-GaN layers on the $(\bar{1}\bar{1}2\bar{2})$ and $(\bar{1}\bar{1}0\bar{1})$ facets; the p-GaN layer on the $(\bar{1}\bar{1}0\bar{1})$ facet shows brighter SEM contrast than that on the $(\bar{1}\bar{1}2\bar{2})$ facet, as shown in Fig. 4.16(a). To discuss quantitatively, Fig. 4.16(c) displays SE intensity profiles along the pn-junction on the $(\bar{1}\bar{1}2\bar{2})$ and $(\bar{1}\bar{1}0\bar{1})$ facets indicated in 4.16(a). As

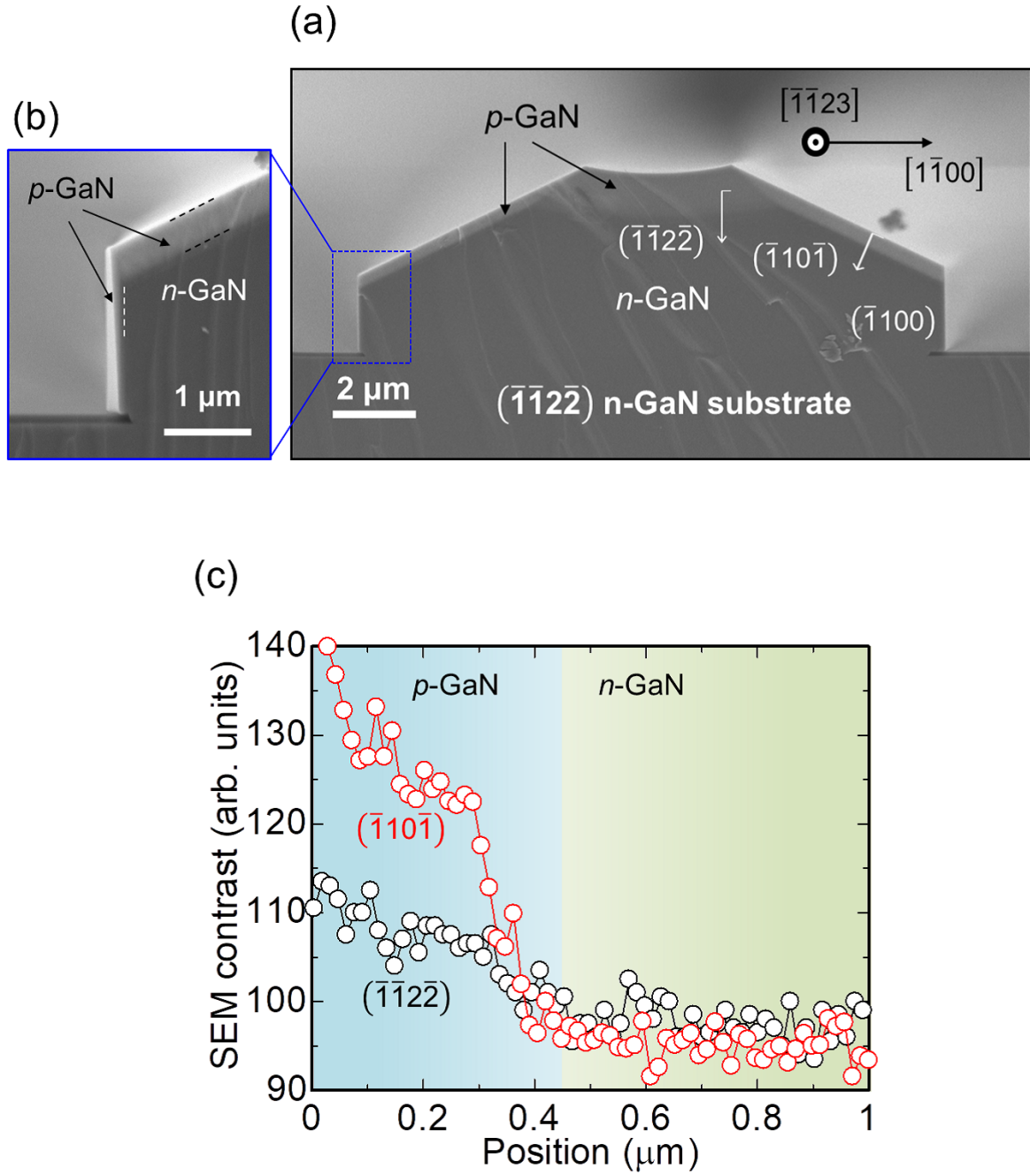


Figure 4.16: Cross-sectional SEM images of (a) a fabricated 3D LED structure on $(\bar{1}\bar{1}2\bar{2})$ and (b) the magified inclined $(\bar{1}10\bar{1})$ and side $(\bar{1}100)$ facets. The II/III ratio during the p-GaN growth is 1.0e-3. (c) SE intensity profiles of pn-junction on the $(\bar{1}\bar{1}2\bar{2})$ and $(\bar{1}10\bar{1})$ facets.

expected, the p-GaN layer on the $(\bar{1}10\bar{1})$ facet emits the larger SE intensity than that on the $(\bar{1}\bar{1}2\bar{2})$, suggesting different p-type conductivity such as carrier concentrations. Previously, Alugubelli *et al.* confirmed higher SE emission intensities from the p-GaN with higher Mg concentrations [28]. Therefore, one possible reason for the higher SE intensity from the $(\bar{1}10\bar{1})$ facet may also be due to the higher Mg concentrations. Indeed, Cruz *et al.* investigated Mg concentrations of Mg-doped GaN layers on various crystallographic orientations and revealed that $(\bar{1}10\bar{1})$ has higher Mg incorporation efficiency than $(\bar{1}\bar{1}2\bar{2})$ [6]. Additionally, they also reported that both the $(\bar{1}10\bar{1})$ and $(\bar{1}\bar{1}2\bar{2})$ show lower incorporation of O [6], which is a well-known compensator in p-type GaN. These findings are consistent with our results. The p-GaN layer on the $(\bar{1}100)$ side facet also exhibits bright SEM contrast, but the thinner thickness disturbs quantitatively comparing the SE intensity with the other facets. This is because, the edge effect in a SEM observation also enhances SE emission intensity in addition to the p-type doping.

4.3.3 Dopant profiling using SPM-based techniques

SEM contrast is an attractive way of monitoring the dopant distribution, but the quantitative analysis is quite difficult: Namely, it is very challenging to make the SE emission intensity profiling correspond to the carrier density profiling. One of the alternative ways to overcome this issue is SPM related techniques: the electrically enabled variants of SPM can probe sample electronic properties with the nanometer spatial resolution. Particularly, scanning microwave microscopy (SMM) combined with scanning nonlinear dielectric microscopy (SNDM) are very powerful tools to characterize the nanoscale profiling of dopant concentration. It should be emphasized that, unlike SEM observations, SMM and SNDM can evaluate the local electronic properties (local capacitance) using the conductive atomic force microscopy (AFM) tips. SMM can contribute to the quantitative determination of dopant concentrations in semiconductor structures, whereas SNDM can identify the dopant type (p or n) and the dopant distribution can be imaged over a wide range from very low to very high carrier concentrations ($10^{14} \sim 10^{20} \text{ cm}^{-3}$). The basic principles of both the SMM and SNDM measurements are described in the following.

SMM and SNDM measurements

Schematic of the SMM and SNDM systems are illustrated in Figs. 4.17(a) and 4.17(b), respectively. In SMM [Figs. 4.17(a)], the AFM tip, which is in contact with the sample surface, is connected to the performance network analyzer (PNA) with

a typical impedance of $50\ \Omega$. The microwave signal generated by the PNA is applied to the sample, and the amount of signal is reflected from the sample due to the impedance mismatch between the sample impedance and the transmission line impedance; in turns, this mismatch depends on the material properties of the sample, which is, in our case, the capacitance of the metal-oxide-semiconductor (MOS) structure underneath the AFM tip. You can find the doping concentration dependence of MOS capacitance in standard textbooks. A $50\ \Omega$ shunt resistor in conjunction with a half-wavelength coaxial resonator is used to realize the higher sensitivity, which is decreased if the sample impedance is too far from $50\ \Omega$. Further details can be found elsewhere [38–43]. SMM has already been applied to a wide range of materials including Si [39, 40], GaAs [43], and GaN [42].

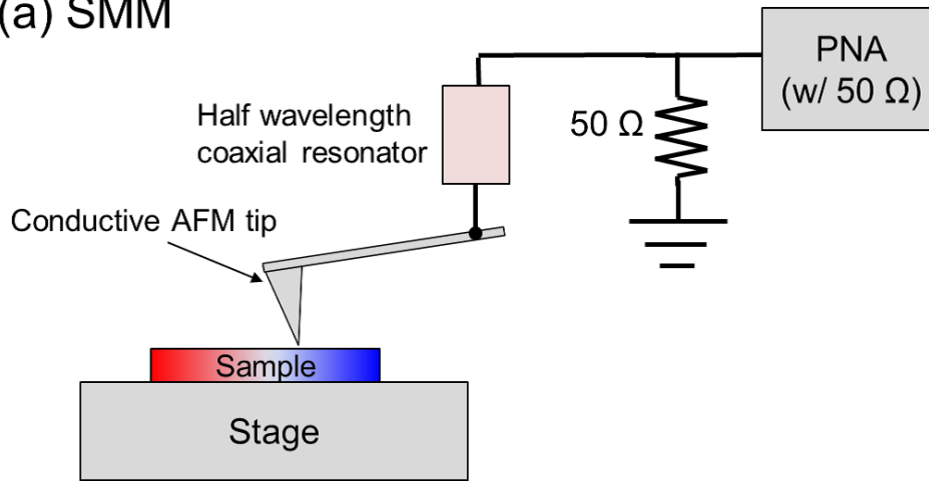
In SNDM [Figs. 4.17(b)], the capacitor sensor, called the SNDM probe, consists of a GHz-range free-running active LC oscillator and conductive AFM tip. When the tip touches the sample surface, the oscillating frequency f_0 of the SNDM probe is determined by the static capacitance C_0 and the tip-sample capacitance $C_s(t)$. The external voltage V applied between the sample and tip modulates the $C_s(t)$, which causes modulation of f_0 . The oscillating frequency deviation $\Delta f(t)$ is proportional to the tip-sample capacitance variation $\Delta C_s(t)$, and the relation is given by

$$\Delta f(t) \propto \Delta C_s(t) = V_{s,0} \left. \frac{\partial C}{\partial V} \right|_0 \cos \omega_s t. \quad (4.1)$$

Considering measurements of semiconductors, the tip and sample form a MOS structure, similar with the SMM configuration. Therefore, $C_s(t)$ is considered to be the MOS capacitance. Then, the typical C_s - V properties for the p- and n-type semiconductors can be illustrated in the inset of Fig. 4.17(b), and the slope of the C_s - V curve, $\partial C_s / \partial V$, is negative for n-type and positive for p-type, giving us the information on the carrier polarity. It should be noted that the extremely high sensitivity of $10^{-22}\text{F}/\sqrt{\text{Hz}}$ enables us to apply this technique to compound semiconductors with much lower signal level than Si. More details can be found in Refs. [44–47]. Wide-bandgap compound semiconductors of SiC or (Al, Ga)N systems have been evaluated by SNDM: the dopant distribution and depletion layer in a cross-section of a SiC double diffused MOSFET [45] or the carrier and polarization distribution in the AlGaIn/GaN heterostructure [46].

Let us describe experimental details in this study. The SMM and SNDM measurements were performed by MST. To evaluate carrier densities quantitatively by SMM, a standard sample with already-known carrier densities were preliminarily prepared. The standard sample was grown on (0001) sapphire substrates, and the structure consists

(a) SMM



(b) SNDM

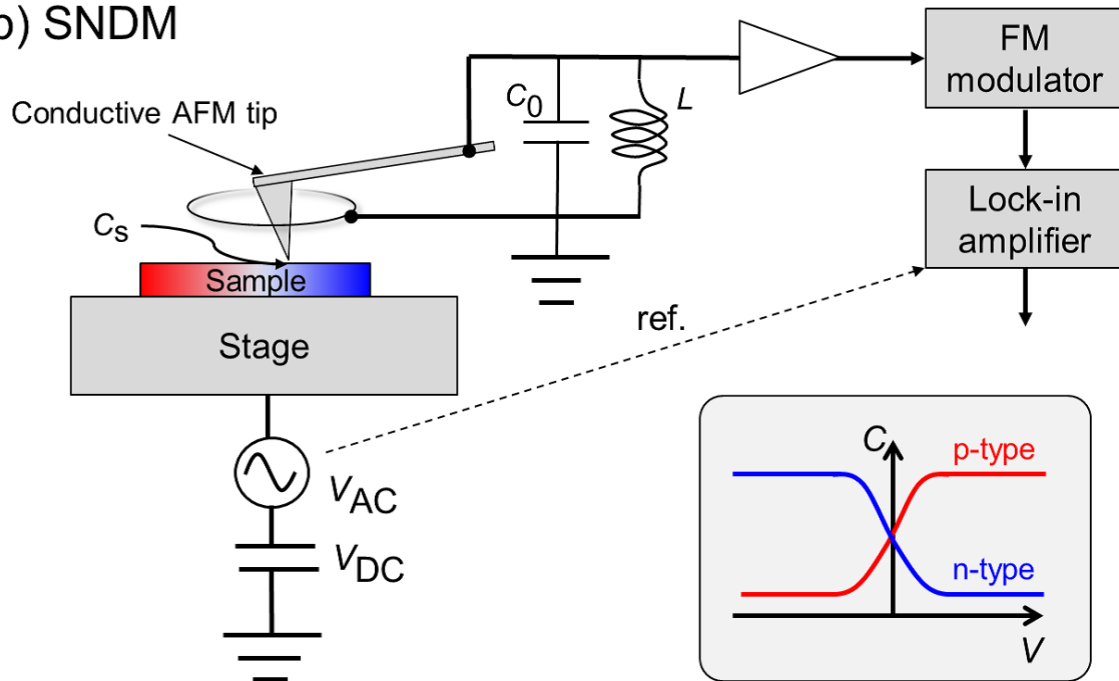


Figure 4.17: Schematic diagrams of (a) SMM and (b) SNDM systems.

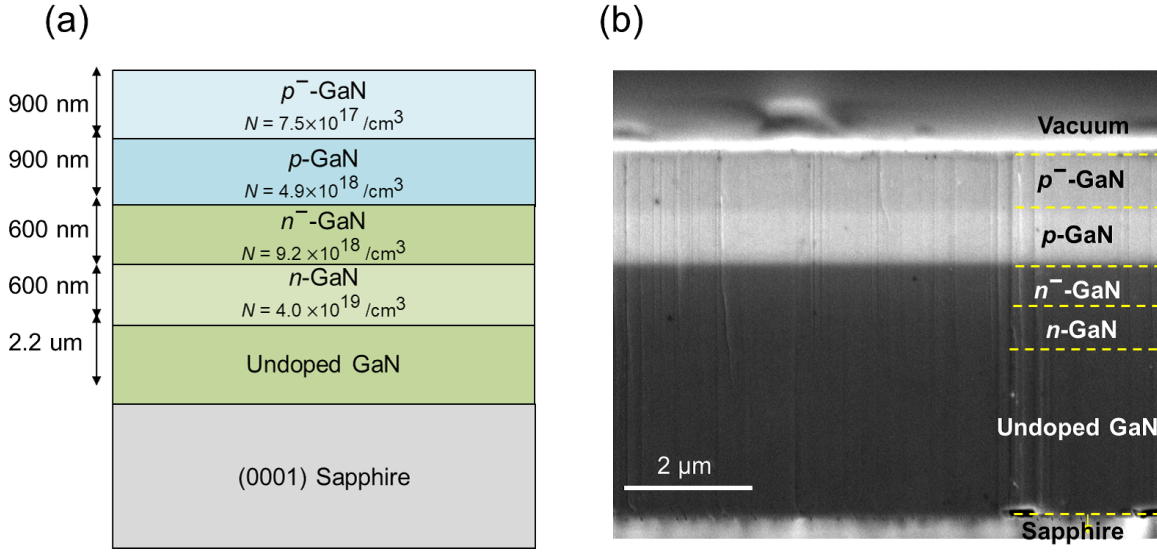


Figure 4.18: (a) Schematic cross section and (b) cross-sectional SEM image of the standard planar sample for carrier density calibration in SMM measurement.

of undoped GaN/ n -GaN/ n^- -GaN/ p -GaN/ p^- -GaN, as shown in Fig. 4.18. The n - and p -type carrier densities were controlled by the Si flow rates and Mg/Ga flow ratios, respectively. The carrier density of each layer was in advance estimated by Hall measurements, using the separately prepared samples composed of the respective n -type or p -type GaN layers on sapphire substrates. The cross-sectional SEM image in Fig. 4.18(b) shows the p^- -GaN layer with darker SEM contrast than the p -GaN layer, which is consistent with the previous discussion. We note that a clean surface is crucial for distinguishing slight dopant contrast; thus, the sample was cleaned by SPM solution ($\text{H}_2\text{SO}_4 : \text{H}_2\text{O}_2 = 3 : 1$) and dilute HF before the SEM measurement.

results and discussions

Figure 4.19 shows (a) carrier polarity and (b) carrier density distributions of the 3D LED structure on $(\bar{1}\bar{1}2\bar{2})$ estimated by SNDM and SMM, respectively. The growth condition of the 3D LED structure was the same as Fig. 4.13. From Fig. 4.19(a), the p - and n -type carrier polarity is successfully visualized in the cross-section by SNDM measurement. The typical carrier densities extracted from the SMM results are also indicated. Here, the carrier density of the p -GaN layer was calibrated from the p -GaN and p^- -GaN layers of the standard sample, whereas that of the n -GaN layer was done from the n -GaN and n^- -GaN layers of the standard sample. The sufficient carrier densities in the order of 10^{18} cm^{-3} and 10^{19} cm^{-3} are confirmed in the p - and n -GaN

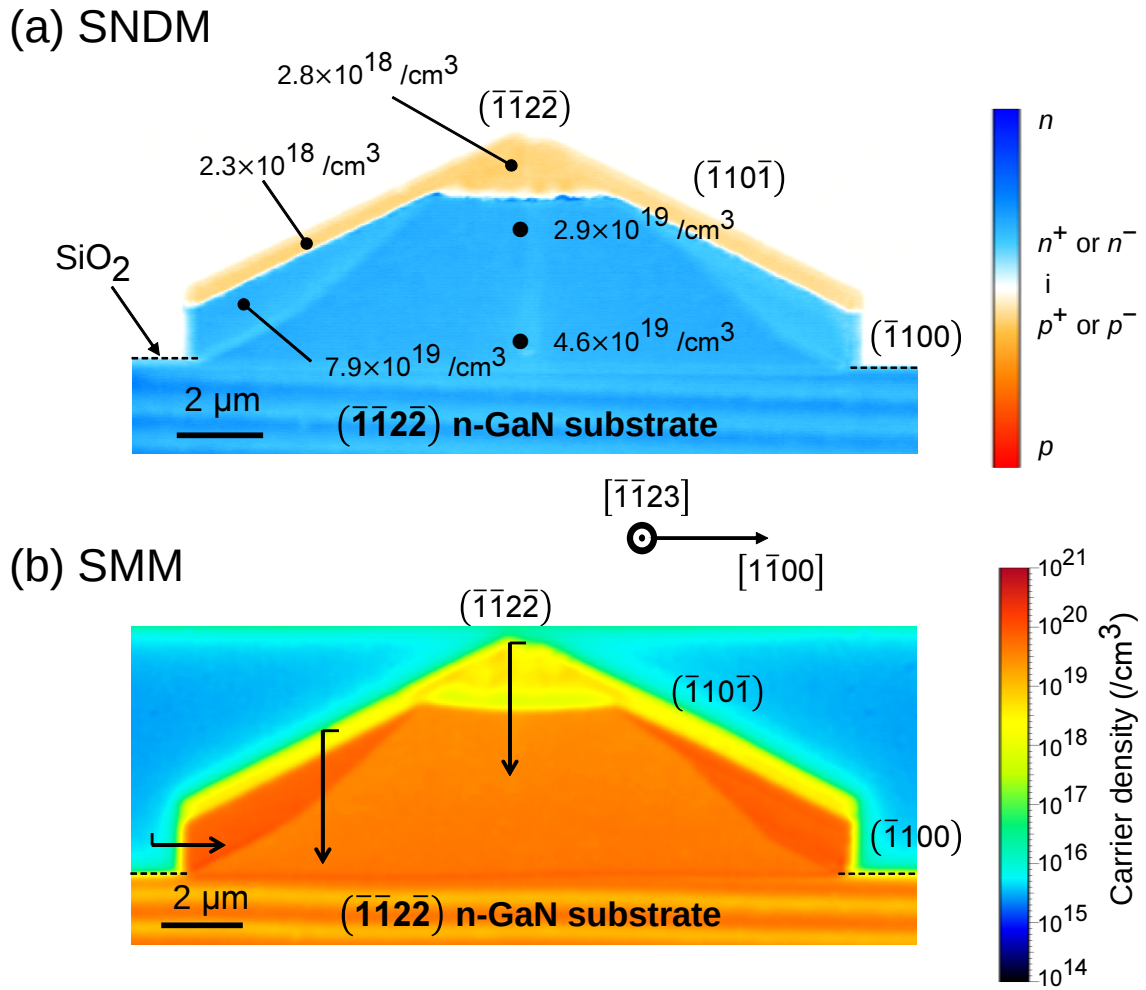


Figure 4.19: (a) Carrier polarity and (b) carrier density distributions evaluated by SNDM and SMM measurements, respectively. It should be noted that the carrier density in (b) was calibrated from the n- and n⁻-GaN of the standard sample.

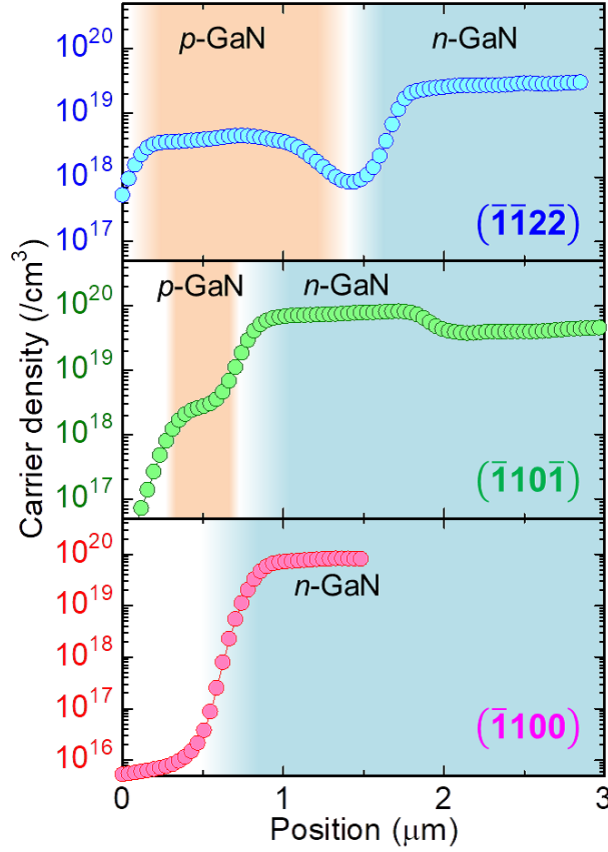


Figure 4.20: A carrier density profile on each facet of a 3D LED structure on $(\bar{1}\bar{1}2\bar{2})$. The profiling positions are indicated in Fig. 4.19(b).

layers, respectively. Additionally, the $(\bar{1}\bar{1}2\bar{2})$ and $\{\bar{1}10\bar{1}\}$ facets have insulating areas between the p- and n-type layers, which correspond to the depletion regions. On the other hand, p-GaN layer is not found on the $\{\bar{1}100\}$ side facet, and that disagrees with the SEM observation. One of the plausible reasons is that the SNDM measurement misjudged the carrier polarity of the Mg-doped GaN layer on the $\{\bar{1}100\}$ side facet due to the spatial resolution comparable to the small p-GaN thickness (< 100 nm). The spatial resolution of SNDM is generally determined by the curvature radius of the AFM tip ($25 \sim 40$ nm), the pixel number of the data (512×256), and the depletion layer expansion under the tip (several tens to hundreds nanometers). The other possible reason is that the Mg-doped GaN layer on the $\{\bar{1}100\}$ side facet does not have p-type conductivity due to the poor doping condition. Indeed, as will be shown in Appendix C, the 3D LED on $(\bar{1}\bar{1}2\bar{2})$, in some cases, shows much amount of the leakage current and

simultaneously indicates the violet emission probably originated from the side facet. In any case, a future experimental study is expected toward more efficient Mg doping and/or wider p-GaN thickness on the side facet.

The carrier density mapping shown in Fig 4.19(b) indicates the distinct spatial distribution of the n-type carrier density. Here, the carrier density was calibrated from the n- and n⁻-GaN of the standard sample, and it should be noted that no remarkable difference from the calibration using the p- and p⁻-GaN was found. The carrier density of $\sim 7.9 \times 10^{19} \text{ cm}^{-3}$ around the $\{\bar{1}10\bar{1}\}$ facet is higher than that of the other n-GaN area ($2.9 \sim 4.6 \times 10^{19} \text{ cm}^{-3}$). The higher carrier density area is well accounted for by the shape evolution of the 3D n-GaN structure [see Fig. 2.10(b)], thus the distinct carrier density distribution could be originated from the higher impurity incorporation (probably, Si and/or O as a donor) of the $\{\bar{1}10\bar{1}\}$ plane. Here, it is noteworthy that the SEM contrast in n-GaN layer is insufficient to observe such a precise difference, and the SMM and SNDM measurements give us the deeper insight into the dopant distribution. The carrier density profiling on each facet was extracted from the designated position in Fig. 4.19(b), and the results are summarized in Fig. 4.20. As a noticeable result, the p-type carrier density on the ($\bar{1}\bar{1}2\bar{2}$) facet indicates a slight delay near the pn junction. This behavior is considered to be related to the Mg doping profile shown in Fig. 4.9. Finally, the above discussions lead us to conclude that SMM and SNDM are quite useful tools for characterizing spatial distributions of the 3D faceted InGaN LED structures with sub- μm scale, and, to the best of our knowledge, this work is the first demonstration of SMM and SNDM measurements to the micro-structured InGaN LEDs.

4.3.4 Optical characterizations

Optical property of the fabricated 3D LED structures on ($\bar{1}\bar{1}2\bar{2}$) with the p-GaN growth condition of II/III ratio = 1.4e-3 (Fig. 4.15) was accessed by photoluminescence (PL) spectroscopy at RT. The PL system was the same as Fig. 3.3. The excitation source was a He-Cd laser (325 nm) with an excitation power density of $\sim 31.8 \text{ W/cm}^2$. Figure 4.21(a) shows the PL spectrum. As a reference, a simultaneously grown ($\bar{1}\bar{1}2\bar{2}$) planar LED was also evaluated. The planar LED exhibits GaN band-edge emission (366 nm), DAP emission ($\sim 380 \text{ nm}$), and InGaN QW emission ($\sim 480 \text{ nm}$). On the other hand, in addition to the GaN band-edge and DAP emissions, the emission spectrum of the 3D LED structure is composed of the $\sim 410 \text{ nm}$ emission and broad emission at around 570 nm. From the previous study in chapter 3 and the cathodoluminescence spectroscopy,

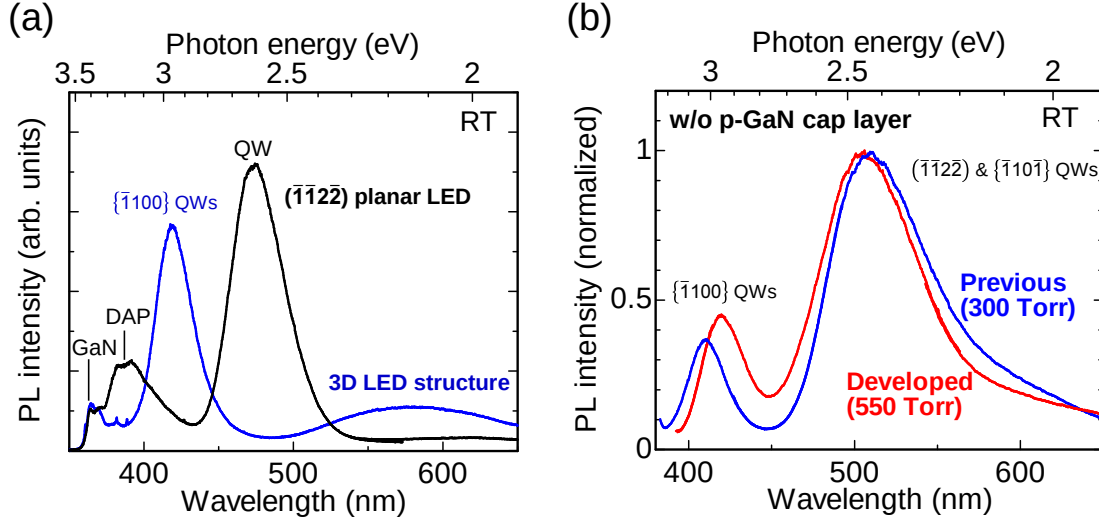


Figure 4.21: (a) PL spectra of 3D LED structures on $(\bar{1}\bar{1}2\bar{2})$ and simultaneously grown $(\bar{1}\bar{1}2\bar{2})$ planar LED. The II/III ratio of the p-GaN layer is 1.4×10^{-3} . (b) PL spectra of 3D InGaN QWs grown under the growth pressures of 300 Torr and 550 Torr.

the ~ 410 nm emission was attributed to the side $(\bar{1}\bar{1}00)$ facet QWs. The broad emission peak at around 570 nm is due to the Si-doped 3D n-GaN and/or n-GaN substrate. Here, the longer wavelength emission from the top $(\bar{1}\bar{1}2\bar{2})$ and inclined $(\bar{1}\bar{1}0\bar{1})$ facet QWs is not observed. To confirm that, 3D InGaN QWs without a p-GaN layer was fabricated. It should be noted that the QW growth pressure was increased from 300 Torr in chapter 3 to 550 Torr in this chapter. Figure 4.21(a) compares emission spectra from the 3D InGaN QWs grown under 300 Torr and 550 Torr. The 3D QWs grown under 550 Torr has a similar polychromatic emission spectrum with that of those grown under 300 Torr. We additionally note that the emission intensities are also similar to each other. Thus, the growth pressure of InGaN QWs is found not to affect the emission characteristics severely. The reason why the longer wavelength emission is not observed in Fig. 4.21(a) would be attributed to the absorption of excitation light by the p-GaN layer.

4.4 Summary

MOVPE growth conditions of polar-plane-free faceted 3D LED structures on $(\bar{1}\bar{1}2\bar{2})$ were explored toward the electrically injected devices. In advance, doping conditions for $(\bar{1}\bar{1}2\bar{2})$ planar LEDs were investigated, expecting difficulty in obtaining proper doping profiles on the $(\bar{1}\bar{1}2\bar{2})$ crystallographic orientation. We firstly revealed that the

doping conditions providing fairly good (11 $\bar{2}2$) LEDs cause poor doping profiles in the ($\bar{1}\bar{1}2\bar{2}$) LEDs. Therefore, using the SIMS profiles for a clue, the doping conditions were optimized by adjusting growth pressure and Mg/Ga flow ratios. The established doping condition lead to the ($\bar{1}\bar{1}2\bar{2}$) LED emitting bright EL through the improvement of the doping profiles. It was found that both growth pressures and Mg flow rates of p-GaN and QW layers play important roles in the doping profiles. Finally, the established LED growth condition for (11 $\bar{2}2$) plane was expanded to the growth of polar-plane-free faceted 3D LED structures on ($\bar{1}\bar{1}2\bar{2}$). Cross-sectional SEM observations provided spatial dopant distribution in the 3D LED structures, and pn-junction formation on each facet was confirmed. Furthermore, as an alternative way, SMM combined with SNDM was demonstrated to successfully yield the deeper insight into the local electrical properties.

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Chapter 5

Polar-Plane-Free Faceted 3D InGaN LEDs

5.1 Introduction

The studies in the previous chapters motivate us to realize electrically driven polar-plane-free 3D LEDs. As an introduction, so far reported device performances of 3D faceted InGaN LEDs are briefly reviewed. The relevant background and essential issues in this research field are provided.

There are some reports on device operations based on several kinds of 3D InGaN LEDs [1–17]. Representative 3D LED structures such as nanocolumn, microrod, multifacet-LED are summarized in Fig. 5.1. It is noted that some of the 3D LEDs are aiming at superior “monochromatic” light emitters. Although the results show some promise, the basic device behaviors, *e.g.* diode properties or emission efficiencies, is still in its infancy. One of the reasons is that device processes on highly 3D structures sometimes cause obstacles to appropriate electrode formation, which results in degrading device process tolerance and disturbing the appropriate diode performance. Another possible reason is the difficulty of realizing efficient p-type doping in the 3D structures. For example, Rishinaramangalam *et al.* reported that Si and O impurities from the SiO₂ mask diffuse into the selectively-grown epi-layer, and compensate Mg acceptors, preventing efficient p-type doping [17]. Consequently, the severe reverse leakage current at the reverse bias voltage was observed. Unestablished fabrication techniques degrade device performance of these 3D LEDs compared to conventional planar LEDs. As another example, Schimpke *et al.* also observed a minor amount of leakage current under reverse bias and a significant series resistance under the forward bias of microrod

(MR) LEDs [16]. The external quantum efficiency (EQE) of the MR LEDs reached 3% even in the blue region. The conventional planar InGaN LED emitting blue has much higher EQE values around 80%, as shown in Fig. 1.3. In addition, the electroluminescence (EL) images display spatially-distributed spot-like emissions, probably due to the poor device quality. The inferior current-voltage (I - V) characteristics have also been reported by several research groups [5, 8, 10]. According to this situation, very limited papers have reported EQE values of the 3D LEDs; to the best of our knowledge, $\sim 3\%$ by Schimpke *et al.* [16] and $\sim 8\%$ (estimation) by Nami *et al.* [18]. 3D faceted InGaN LEDs require numerous efforts in the fabrication techniques for further improvements.

Here, let us note device performance of the 3D LEDs on (0001) fabricated in our laboratory. The typical EL properties can be found in elsewhere [1, 7]. Mr. K. Hayashi evaluated EQE values of the fabricated LEDs using an integrating sphere and a planar LED with an already-known EQE for the reference, in his master's thesis work. The estimated EQE values were in the range of 0.106~0.78%. The diode performances were the turn-on voltages of 2.9~3.3 V and the series resistances of 325~360 Ω , depending on chips or structures. (In some cases, large leakage currents were also observed as will be shown later.) The higher turn-on voltages were considered to be due to the inter-facet and in-chip distributions of Mg concentrations. The higher series resistances were attributed to the higher series resistances of the 3D n-GaN without Si doping and unoptimized thermal treatments of n- and p-contact electrodes.

In chapter 4, optimal doping conditions of ($\bar{1}\bar{1}2\bar{2}$) planar LEDs have been established, which successfully yield pn-junction formation on each facet in the polar-plane-free faceted 3D structures. In this chapter, device processes toward the electrically injected operations are investigated. We optimize the device processes to meet our polar-plane-free faceted 3D LEDs. Both process conditions of vacuum evaporation and photolithography techniques are examined. After that, we characterize fundamental device properties of the polar-plane-free faceted 3D LEDs. Clear rectifying properties and broadband EL from the fabricated LEDs are demonstrated.

5.2 Device Processes

5.2.1 Experiments

3D LED structures were grown on ($\bar{1}\bar{1}2\bar{2}$) GaN substrates. The growth condition was the same as the established condition in Sec. 4.2. LED device design of the polar-plane-free faceted 3D LEDs is schematically illustrated in Fig. 5.2. Vertical LED structure

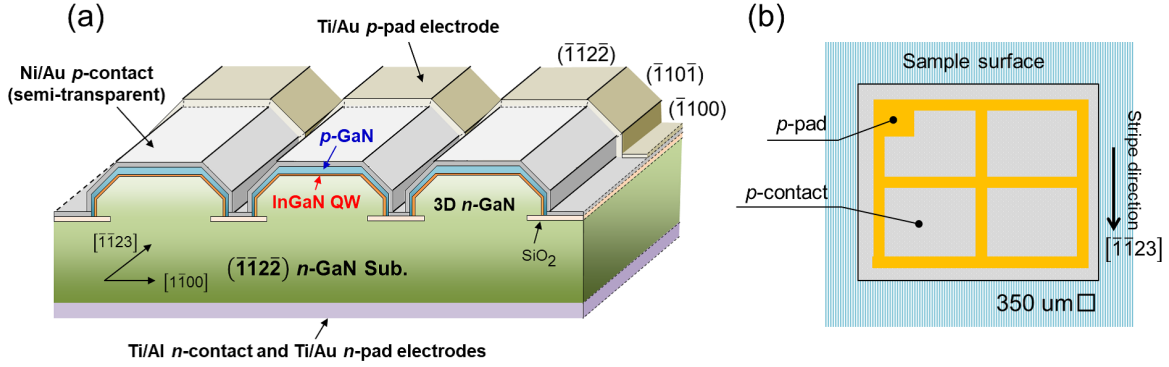


Figure 5.2: A LED design of polar-plane-free faceted 3D LEDs. (a) Cross-sectional view and (b) surface view.

was employed utilizing n-type conductivity of the Si-doped $(\bar{1}\bar{1}2\bar{2})$ GaN substrates. This also allows us to reduce much effort for complicated device processes. The removal of remaining AlN past on the sample backside and HCl treatment was performed before device processes in the same manner as described in Sec. 4.2. Then, Ni (5 nm)/Au (10 nm) electrodes followed by Ti (50 nm)/Au (200 nm) p-pad electrodes were formed on the sample surfaces by photolithography and vacuum evaporation. Finally, Ti (30 nm)/Al (100 nm) n-contact and Ti (50 nm)/Al (100 nm) n-pad electrodes were deposited from the sample backsides. Thermal treatments for these electrodes were the same as Sec. 4.2. The p-contact and p-pad patterns are shown in Fig. 5.2(b). The chip size was 350 μm^2 .

Here, we note that a conventional lateral LED design was at first fabricated, but the fabricated LEDs did not work well. A part of the reasons was attributed to the nonuniform photoresists and deposited metals, as described in the following. Besides them, the processes for lateral LEDs require mesa etching processing by reactive ion etching (RIE) technique. Such a complicated device process would be unfavorable in our case.

5.2.2 Electrode formation

Firstly, device processes with the same process condition as planar films were conducted for the 3D LED structures. As a result, we found undesirable electrode separation after the processes, as shown in Figure 5.3(a), where a scanning electron microscopy (SEM) image of the sample surface after the device processes is presented. The dotted lines indicate the designed pattern of the p-pad electrode. It is found that the p-pad

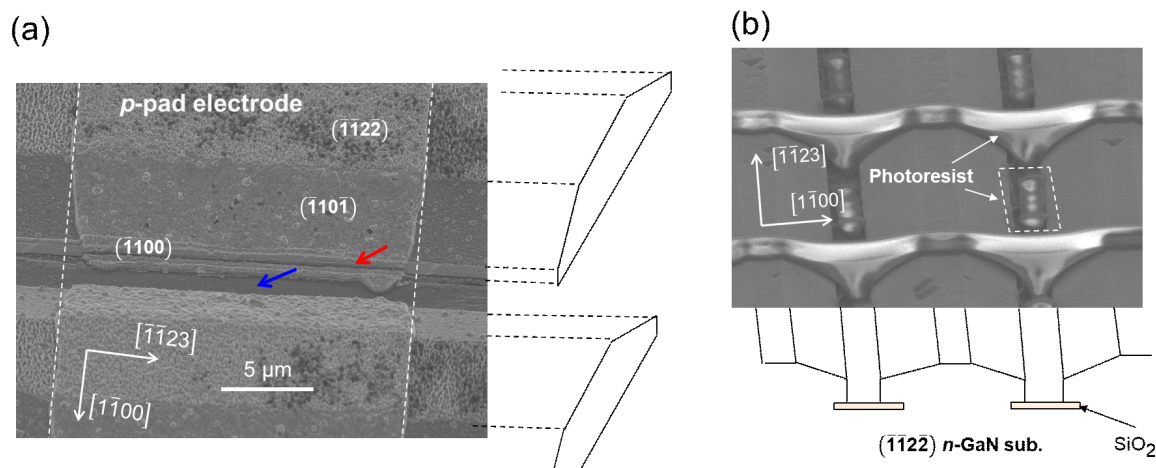


Figure 5.3: Bird's-eye view SEM images of (a) an initially fabricated 3D LED and (b) 3D LED structures after photolithography.

electrode is separated on the SiO₂ mask area designated by the blue arrow. In addition, electrode separation also occurs on the side facet of the 3D structures, as indicated by the red arrow. These undesirable electrode separations apparently disturb the LED device operation. To solve these issues, 3D LED structures during the processes were observed.

Figure 5.3(b) shows a bird's-eye view SEM image of the 3D LED structures after photolithography. The UV exposure time was 5 sec, which was preliminarily optimized for planar films in our laboratory. Striped photoresists are clearly observed, but unremoved photoresist surrounded by a dotted line is also identified on the SiO₂ mask. Therefore, the electrode separation on the SiO₂ mask observed in Fig. 5.3(a) is considered to be caused by the following lift-off process. The unremoved photoresist is caused by the thicker photoresist formation on the SiO₂ mask area and the insufficient UV exposure time. On the other hand, the electrode separation on the side facet is caused by the other process, *i.e.*, vacuum evaporation. In the vacuum evaporation system used in this study, evaporated metal molecules diffuse vertically into the sample surface, which simultaneously means that the side facets are in parallel to the incident direction of diffused metal molecules, resulting in the less deposition on the side facets. Figure 5.4 schematically illustrates the described two situations in photolithography and vacuum evaporation. To solve these issues, the following optimizations were performed: longer UV exposure times for photolithography and oblique-angle deposition for vacuum evaporation. The oblique-angle deposition was performed with a hand-made stage, which enabled us to put samples diagonally.

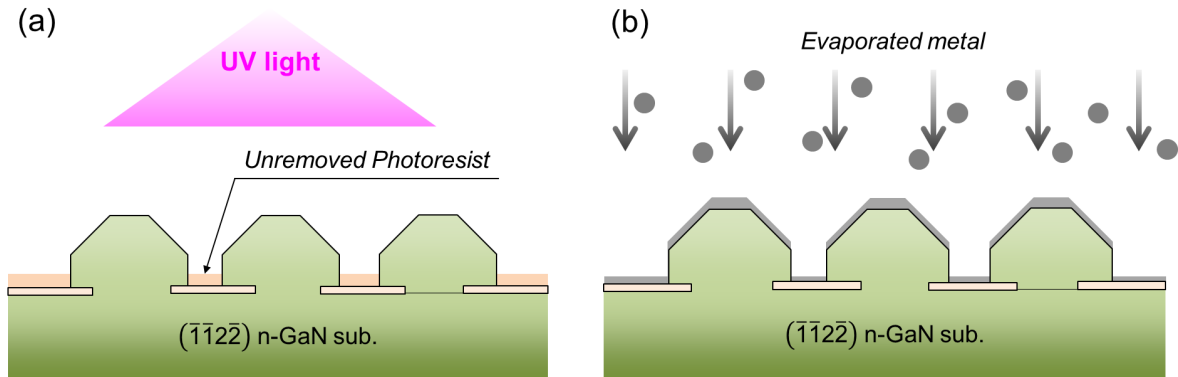


Figure 5.4: Schematic illustrations of device process issues in (a) photolithography and (b) vacuum evaporation.

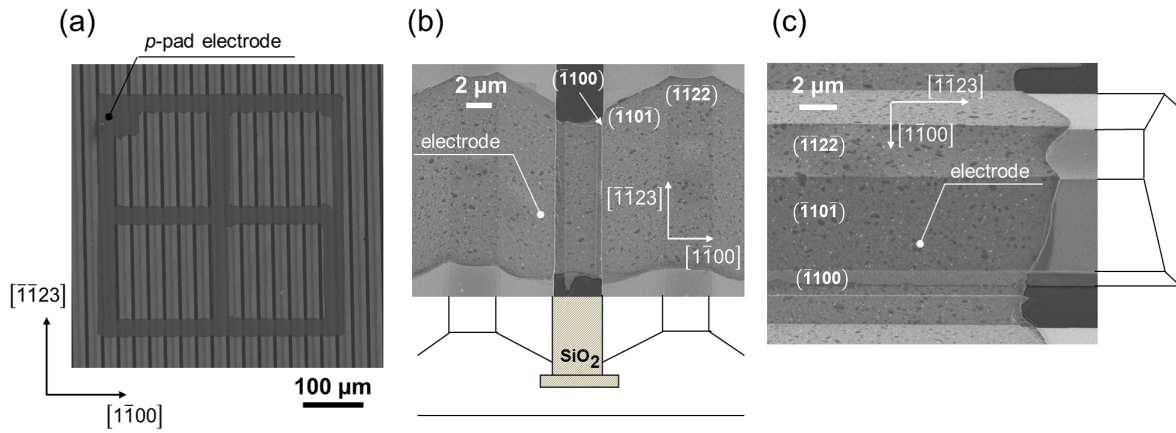


Figure 5.5: SEM images of an optimized 3D LED. (a) Surface view of the 3D LED with a chip size of $350 \mu\text{m}^2$. Magnified SEM image of (b) the SiO_2 mask area and (c) the side facet

Figure 5.5 shows SEM images of the 3D LEDs processed by optimized process conditions. The UV exposure time was lengthened to 11 sec. Figure 5.5(a) clearly indicates p-pad electrode corresponding to the design in Fig. 5.2(b). Then, the magnified SEM images on the SiO₂ mask area in Fig. 5.5(b) and the side facet in Fig. 5.5(c) show no electrode separations. Namely, the longer UV exposure time for photolithography and oblique-angle deposition for vacuum evaporation solved the above-mentioned issues. Here, we note that 3D LEDs on (0001) fabricated in our laboratory also required UV exposure time optimization, but vacuum evaporation condition was the same as the planar films [1], unlike the polar-plane-free faceted 3D LEDs in this study. The reason could be explained by their 3D shapes: The 3D LED structures on (0001) is composed of the top (0001) and inclined (11 $\bar{2}$ 2) facets without side facet [1], whereas the polar-plane-free faceted 3D LED structures have the ($\bar{1}$ 100) side facet.

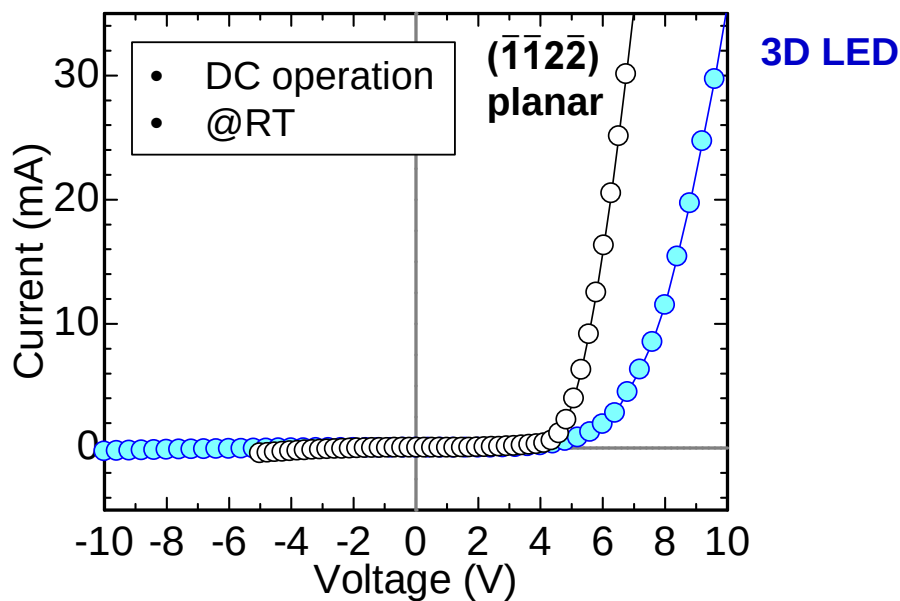
5.3 Device Performance

5.3.1 Fundamental device characteristics

Figure 5.6(a) shows the I - V characteristic of the fabricated polar-plane-free faceted 3D LEDs under direct current (DC) operation at room temperature (RT). For the reference, ($\bar{1}\bar{1}2\bar{2}$) planar LEDs grown under a similar growth condition (~ 300 nm p-GaN thickness) is also shown. Although the turn-on voltage (~ 6 V) of the 3D LED is higher than that of the planar LED (~ 4.5 V), the 3D LED exhibits clear rectifying property. The rectifying behavior indicates pn-junction formation in the 3D LED structures in terms of electrical properties, in addition to the SEM contrast observations in Sec. 4.3.2. Several reasons can be considered for the higher turn-on voltages of the 3D LEDs. Concerning crystallographic orientations, one of the possible reasons is the p-contact resistance contributed from the $\{\bar{1}10\bar{1}\}$ facets, which differs from the planar LED with p-contact on ($\bar{1}\bar{1}2\bar{2}$) plane. The different crystallographic planes may induce different p-contact resistance. On the other hand, electrode formation could also induce the higher turn-on voltage. Precise Ni thickness design (~ 5 nm) is often required to form proper ohmic p-contact on p-GaN. However, as discussed in Sec. 5.2, uniform electrode thickness on the 3D LEDs is quite difficult, which may cause undesirable Ni thickness distribution on the facet surfaces.

Figure 5.6 shows normalized RT-EL spectra from the 3D LED and planar LED at driving currents of DC 20 mA. An example photograph of the emission from the 3D LED at DC 20 mA is also shown. The photoluminescence (PL) spectrum from the 3D

(a)



(b)

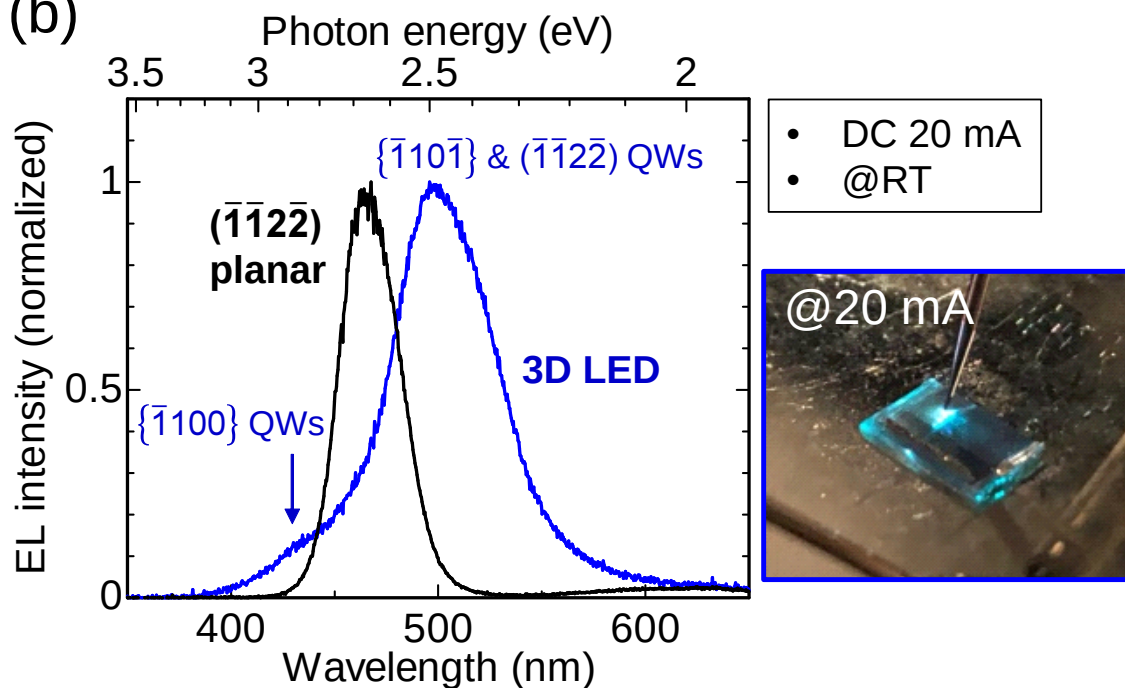


Figure 5.6: (a) *I-V* characteristics and (b) EL spectra from the fabricated 3D LED on $\bar{1}\bar{1}2\bar{2}$ and $\bar{1}\bar{1}2\bar{2}$ planar LED. The inset photograph in (b) shows the 3D LED on $\bar{1}\bar{1}2\bar{2}$ at DC 20 mA.

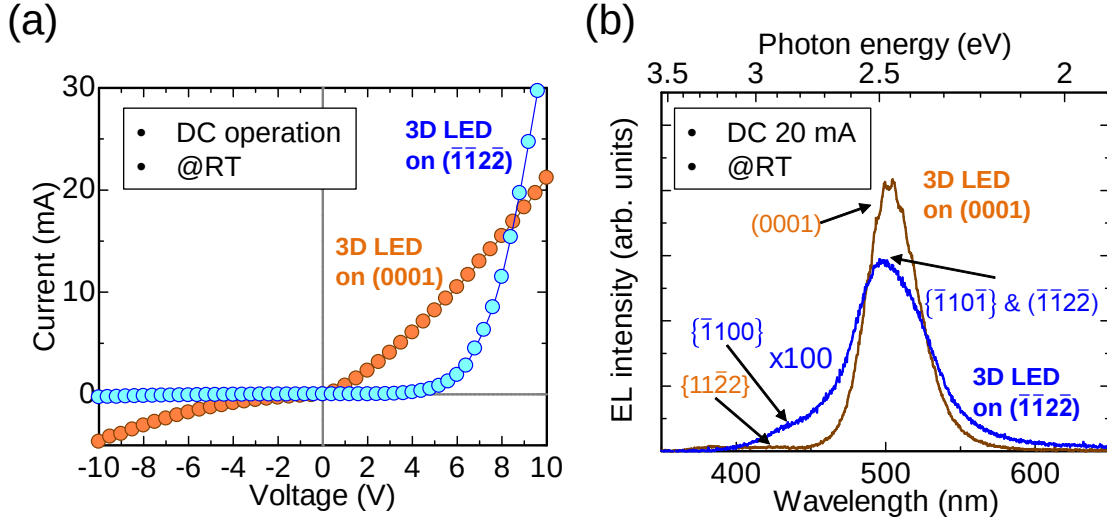


Figure 5.7: Device property comparison between the 3D LEDs on (0001) and $(\bar{1}\bar{1}2\bar{2})$. (a) *I-V* characteristics and (b) EL spectra at DC 20 mA. It should be noted that the EL intensity of the 3D LED on $(\bar{1}\bar{1}2\bar{2})$ is multiplied by 100.

QWs without a p-GaN layer can be found in Sec. 4.3.3. According to this, the main emission peak of the 3D LED locates around 500 nm, which is attributed to the top $(\bar{1}\bar{1}2\bar{2})$ and inclined $\{\bar{1}10\bar{1}\}$ facets. In addition, shorter-wavelength emission from the side $\{\bar{1}100\}$ can be seen as a shoulder at ~ 420 nm. Compared to the spectrum from the planar LEDs, larger EL bandwidth is created by the 3D LEDs due to the polychromatic emission property. These results lead us to conclude that we have successfully achieved electrically driven polar-plane-free faceted 3D LEDs.

Here, let us simply compare the device performance of the 3D LEDs on (0001) and $(\bar{1}\bar{1}2\bar{2})$. The 3D LED on (0001) was fabricated in our laboratory, and the SiO_2 mask design was window width m (μm) and mask width m (μm) of $(w, m) = (15, 5)$, which is the same as the 3D LED on $(\bar{1}\bar{1}2\bar{2})$. The 3D structure consists of the top (0001) facet emitting the longer wavelength component (~ 500 nm) and inclined $\{\bar{1}12\bar{2}\}$ facet emitting the shorter wavelength component (~ 420 nm). The device process of the 3D LEDs on (0001) and $(\bar{1}\bar{1}2\bar{2})$ was almost the same, though the mesa etching process by RIE technique was applied to the 3D LED on (0001), because of the insulating sapphire substrate. The *I-V* characteristics and EL spectra are compared in Figs. 5.7(a) and 5.7(b), respectively. Figure 5.7(a) shows distinct *I-V* characteristics: the 3D LED on (0001) exhibits the poor rectifying property and larger series resistance probably due to the unoptimized doping condition. In particular, the larger series resistance can be attributed to the lower carrier density of the 3D n-GaN layer around $5\sim 8 \times 10^{17}$

cm^{-3} estimated by Raman spectroscopy. As already discussed in Sec. 4.3.3, the carrier density of the 3D n-GaN on $(\bar{1}\bar{1}2\bar{2})$ was evaluated to be in the order of 10^{19} cm^{-3} by SMM measurement. On the other hand, the EL intensity from the 3D LED on (0001) is much larger than that from the 3D LED on $(\bar{1}\bar{1}2\bar{2})$ [Figure 5.7(b)]. This result, at first sight, seems to indicate the superior internal quantum efficiency (IQE) of the 3D LED on (0001), but the light extraction efficiency should also be considered; the sapphire substrate of the 3D LED on (0001) can enhance the light extraction efficiency compared with the GaN substrate of the 3D LED on $(\bar{1}\bar{1}2\bar{2})$ due to the different refractive index n between GaN ($n \simeq 2.5$) and sapphire ($n \simeq 1.8$) at the light wavelength of 500 nm. Therefore, it is difficult to draw a conclusion about the IQE of the 3D LED on (0001) and $(\bar{1}\bar{1}2\bar{2})$ from Fig. 5.7(b), and a future experimental study is expected.

Now, comparing the EL spectrum of the 3D LED on $(\bar{1}\bar{1}2\bar{2})$ with the PL spectrum of the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ in Fig. 4.21(b), the shorter wavelength emission from the side $\{\bar{1}100\}$ facet in EL seems not to be clearly observed. Therefore, the device performance should be improved toward further broadband emission characteristics. Considering the experimental conditions, PL measurements can simultaneously excite all the facet QWs, but the situation is different under the electrically injected conditions (EL). In addition to the emission efficiency of each facet QWs, an amount of injected carriers into the QWs also affects the emission intensity. One of the possible factors is the inadequate p-type doping on $\{\bar{1}100\}$ facets, which may induce the poor injected carriers. The other factor is that the p-contact and p-pad electrode formation on the side facet required elaborate process optimization, expecting that the Ni thickness on the side facet is probably not optimal. However, since the two issues cannot be solved immediately, an alternative approach is required. From such a background, one of the possible ways to enhance the emission intensity from the 3D LEDs on $(\bar{1}\bar{1}2\bar{2})$ is to alter the SiO_2 mask geometry. In the previous reports of 3D LEDs on (0001), it was demonstrated that the emission spectra could be controlled by altering the SiO_2 mask geometry [1, 7, 19]. This is because, the intensity of each emission component depends on the area of the originated facet QWs, and thus, the 3D shape control via altering the mask geometry results in controlling emission spectrum.

5.3.2 Structure-mixed 3D LEDs

As already shown in Fig. 2.7, altering SiO_2 mask geometries affect shapes of 3D GaN structures on $(\bar{1}\bar{1}2\bar{2})$. Therefore, we compare the InGaN QWs overgrown on these 3D GaN structures in an attempt to tailor the emission spectra. Figure 5.8 shows

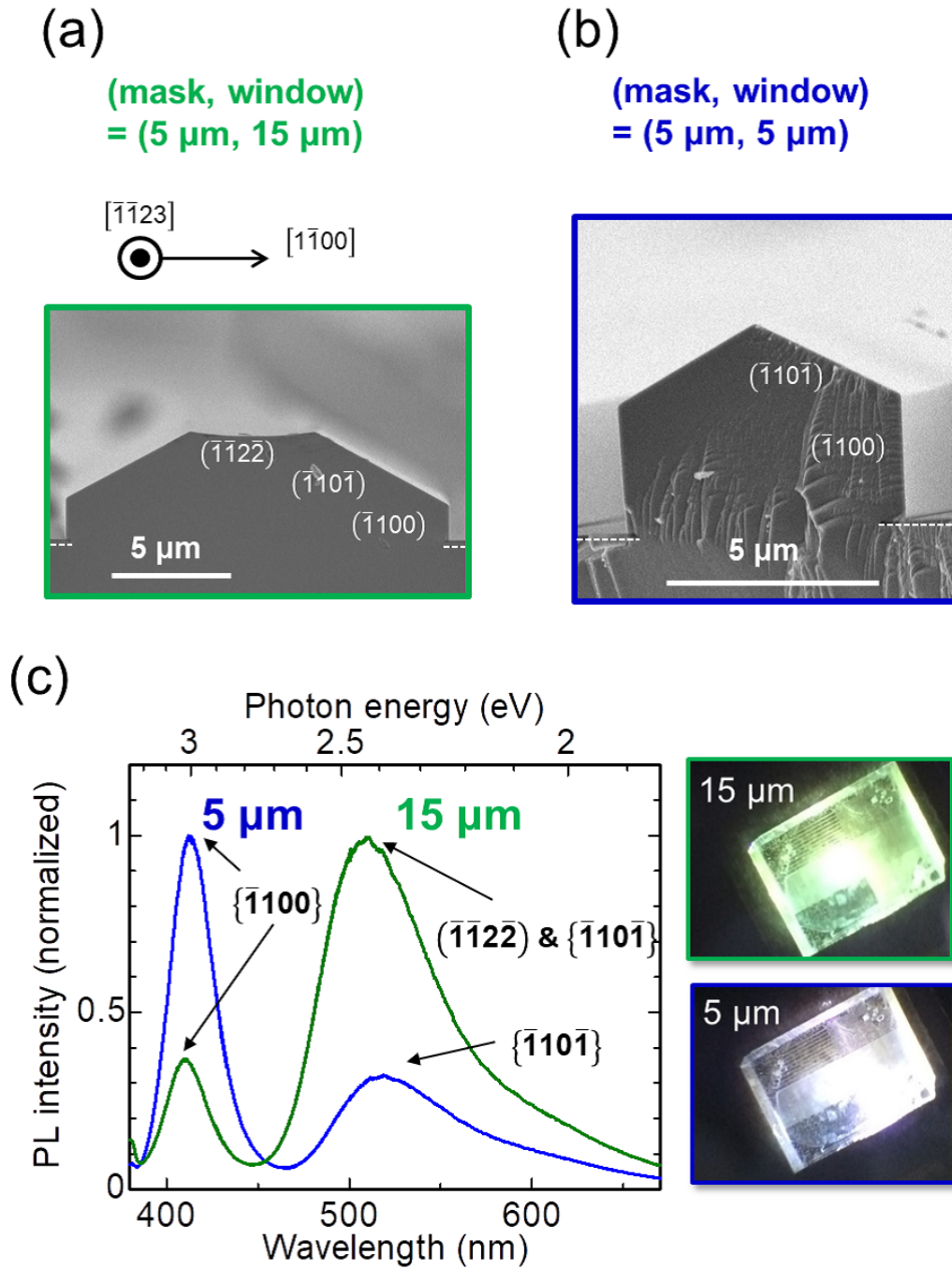


Figure 5.8: Cross-sectional SEM images of 3D QWs on the mask geometries of $(w, m) =$ (a) (15, 5) and (b) (5, 5), and (c) PL spectra from each 3D QWs at RT. The inset photographs show the samples emitting greenish and purplish colors corresponding to the mask geometries.

cross-sectional SEM images of 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ with different mask geometries: The window width w (μm) and mask width m (μm) are $(w, m) = (\text{a}) (15, 5)$ and $(\text{b}) (5, 5)$. The 3D QWs with a window width of $5 \mu\text{m}$ are composed of the inclined $\{\bar{1}10\bar{1}\}$ and the side $\{\bar{1}100\}$ facets. The relative facet width of the side to inclined facets becomes smaller than that of 3D QWs with the window width of $15 \mu\text{m}$ (Fig. 5.8). This observation makes us expect the variation of the relative emission intensity. It should be noted that both the 3D QWs are simultaneously grown under the same growth conditions. As expected, the PL spectra clearly depend on the 3D shapes, as shown in Fig. 5.8(c). For the 3D QWs with a window width of $15 \mu\text{m}$, the emission intensity of the longer wavelength component originated from the top $(\bar{1}\bar{1}2\bar{2})$ and inclined $\{\bar{1}10\bar{1}\}$ facets is larger than that of the shorter wavelength component. On the other hand, the larger emission intensity of the shorter wavelength component can be realized by 3D QWs with the window width of $5 \mu\text{m}$, owing to the shape variation. These results lead us to conclude that the intensity ratio can be tuned by altering the SiO_2 mask geometries. The distinct emission spectra result in apparently different emission colors indicated in the inset photographs of Fig. 5.8(c). These experimental results inspired us to combine these different mask patterns in order to strengthen shorter wavelength emission from the polar-plane-free faceted 3D LEDs.

Figure 5.9(a) shows schematic of pattern mixture, which is composed of one mask width ($5 \mu\text{m}$) and two different window widths ($15 \mu\text{m}$ and $5 \mu\text{m}$). It should be noted that this new period is still smaller than the typical dimension of LEDs ($\sim 300 \mu\text{m}$), and thus, the present proposal is applicable to practical devices. A cross-sectional view of 3D LED structures grown on the mixed pattern depicted in Fig. 5.9(a) was taken by SEM, and a typical image is shown in Fig. 5.9(b). As expected, the 3D structures on window widths of $5 \mu\text{m}$ and $15 \mu\text{m}$ [Figs. 5.8(a) and 5.8(b)] simultaneously appear according to the designed mask pattern. The cross sections in Fig. 5.9(b) were deformed from those in Figs. 5.8(a) and 5.8(b) due to the additional growth of p-GaN layers. SEM contrast caused by p- and n-type conductivity can be confirmed (The detailed discussion on SEM contrast can be found in Sec. 4.3.2).

After the same device processes explained in Sec. 5.2, the device performances were evaluated at RT. Figure 5.10 shows (a) a I - V curve under DC operation and (b) a typical EL spectrum at driving current of 20 mA . The turn-on voltage is still higher, but the clear rectifying property is observed. Compared to the EL spectrum from the previous 3D LED (Fig. 5.6), two emission peaks at around 410 nm and 500 nm are clearly identified. Due to the more broadband emission, the apparent color looks whitish, as shown in the inset photograph of Fig. 5.10(b). Therefore, we have

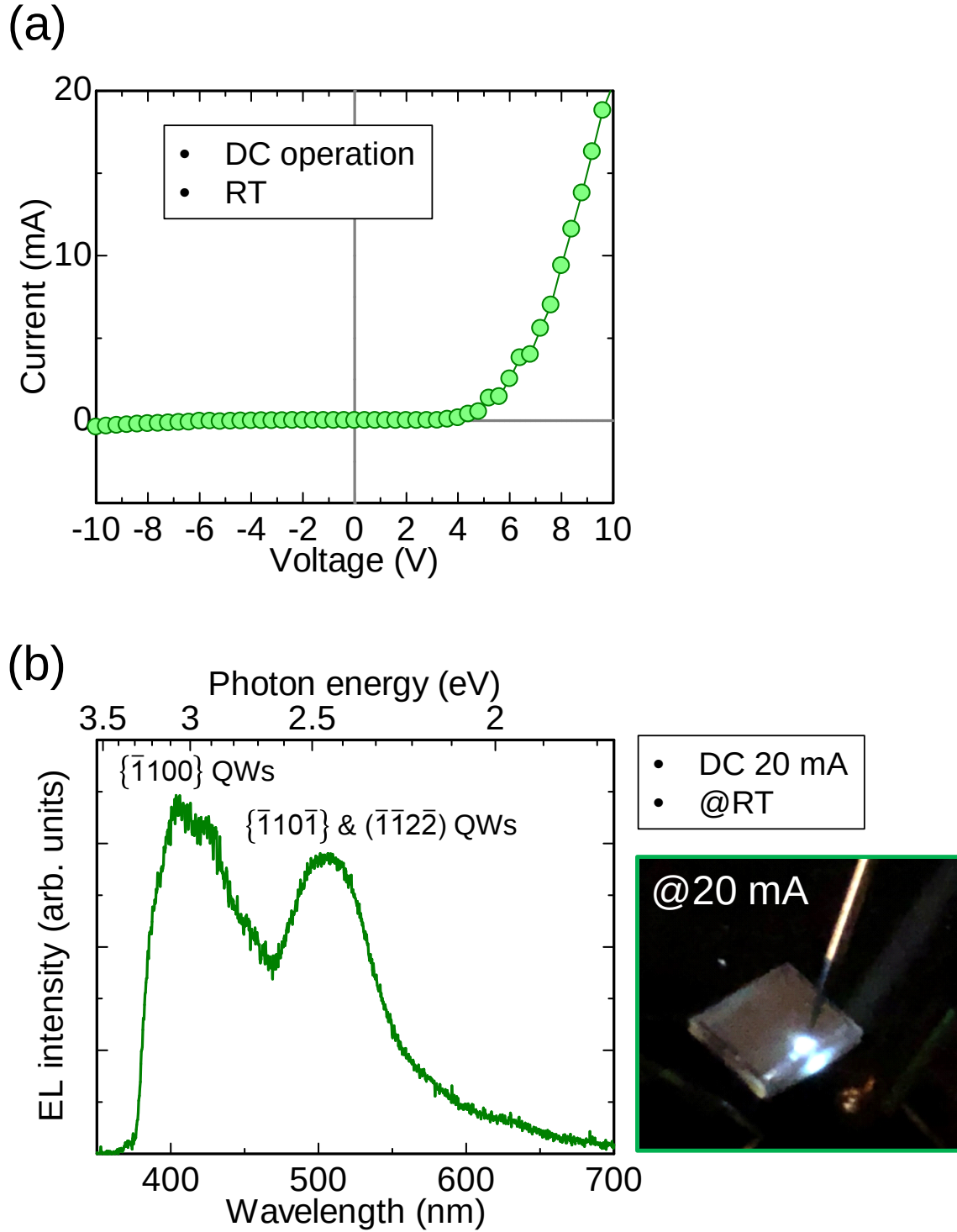


Figure 5.10: (a) I - V characteristic and (b) EL spectrum from the fabricated 3D LED on $(\bar{1}\bar{1}2\bar{2})$ with a pattern mixed SiO_2 mask indicated in Fig. 5.9.

successfully strengthened shorter wavelength emission from the 3D LED on $(\bar{1}\bar{1}2\bar{2})$.

5.3.3 Toward white color syntheses

Finally, we attempted to further longer wavelength emission from the 3D LEDs on the pattern mixture mask, aiming at white color synthesis. The growth temperature of the InGaN QW layer was decreased to increase the In compositions in the same manner as Sec. 3.7. The same growth conditions as the previous samples were applied to the other parts of the epitaxial layers (n- and p-GaN layers). Figure 5.11 displays the device characteristics. The rectifying property becomes worse compared to the previous 3D LEDs (Figs. 5.10 and 5.6), and the larger leakage currents were observed. This variation would be caused by the more defective InGaN QW due to the lower growth temperature. Accordingly, the EL spectrum displayed in Fig. 5.11(b) is quite weak, but the very broad emission, fortunately, synthesizes the white color shown in the inset photograph of Fig. 5.11(b). The very broad spectrum makes it difficult to assign the origins of the emission color component. A future experimental study is required to improve the device performance.

The emission colors of the fabricated 3D LEDs on $(\bar{1}\bar{1}2\bar{2})$ under DC 20 mA are summarized in terms of the Commission Internationale de l'Éclairage (CIE) 1931 chromaticity diagram in Fig. 5.12. As a reference, the emission from the $(\bar{1}\bar{1}2\bar{2})$ planar LED in Fig. 5.6(b) is also shown. The monochromatic emission spectrum from the planar LED is at (0.152, 0.081) near the boundary in the CIE chromaticity diagram. On the other hand, the emission from the 3D LED in Fig. 5.6(b) locates at (0.159, 0.405) far from the boundary and shifts to blueish-green region due to the polychromatic emission. Then, the strengthened shorter wavelength emission of the structure-mixed 3D LED in Fig. 5.10(b) results in the more broadband spectrum, thus the plot moves close to the white color region (blueish-white) at (0.201, 0.281). The pronounced broad emission from the structure-mixed 3D LED aiming at white color synthesis in Fig. 5.11(b) moves more inside the CIE chromaticity diagram, and eventually, the plot locates at (0.325, 0.390) in the white color region. These results show some promise for arbitral pastel color syntheses based on the polar-plane-free faceted InGaN LEDs on $(\bar{1}\bar{1}2\bar{2})$.

5.4 Summary

Polar-plane-free faceted 3D LEDs on $(\bar{1}\bar{1}2\bar{2})$ were fabricated, and the LED device performance was investigated. The device process conditions for planar films were found

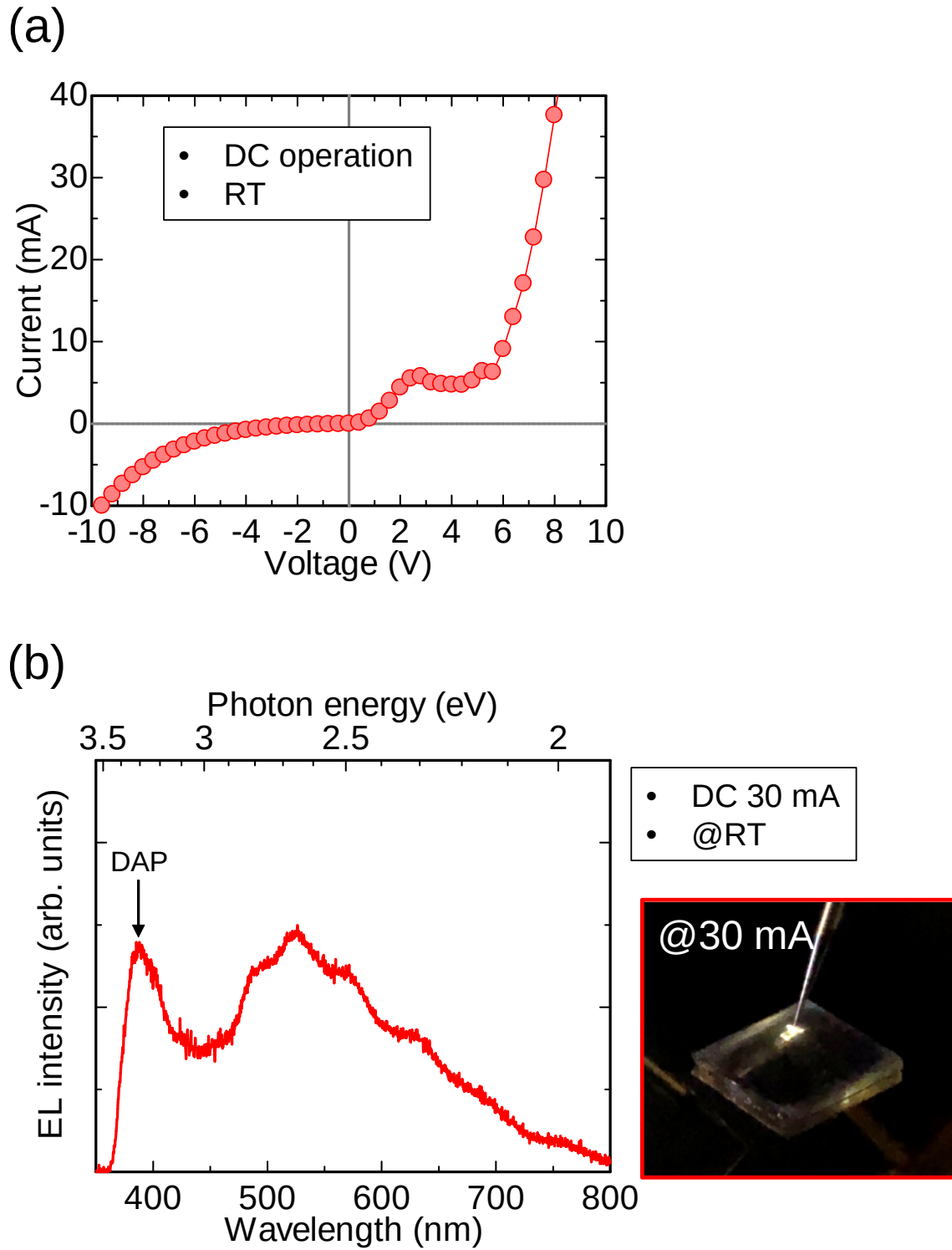


Figure 5.11: (a) I - V characteristic and (b) EL spectrum from the fabricated 3D LED on $(\bar{1}\bar{1}2\bar{2})$ aiming at white color synthesis.

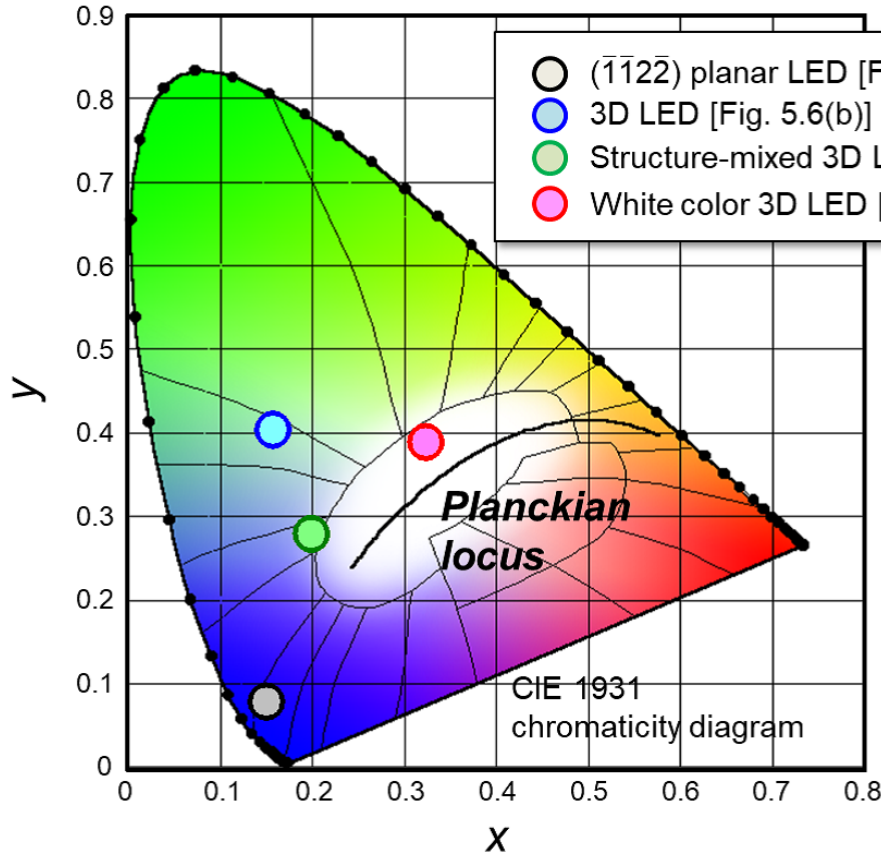


Figure 5.12: CIE chromaticity diagram for the 3D LEDs on $(\bar{1}\bar{1}2\bar{2})$ fabricated in this study. The $(\bar{1}\bar{1}2\bar{2})$ planar LED is also plotted as the reference.

to be unsuitable for the 3D LEDs due to the three-dimensionality, and the conditions were optimized for the 3D LEDs. The developed device processes successfully excluded the electrode separation issue. The 3D LEDs on $(\bar{1}\bar{1}2\bar{2})$ fabricated by the optimized device processes exhibited clear rectifying properties and broad EL spectra compared with the $(\bar{1}\bar{1}2\bar{2})$ planar LED. In addition, the structure-mixed 3D LEDs fabricated by altering the SiO_2 mask geometry showed a well-controlled EL spectrum corresponding to the 3D structures. Finally, white color synthesis based on the 3D LEDs on $(\bar{1}\bar{1}2\bar{2})$ was attempted, and actually, the emission looked whitish color. However, the emission intensity was quite low, indicating that further improvements in all the component efficiencies are necessary. In any case, it is noteworthy that the polar-plane-free faceted 3D LEDs on $(\bar{1}\bar{1}2\bar{2})$ can provide polychromatic emissions under electrically injected operations.

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Chapter 6

Conclusions

This study aimed at exploring the potential of semipolar physics to the 3D faceted InGaN-LEDs with polychromatic emission properties for future advanced light emitters. For this purpose, we investigated three-dimensionally (3D) faceted InGaN-LED structures grown on $\{11\bar{2}2\}$ GaN substrates. This work was particularly devoted to the growth characteristics during metalorganic vapor phase epitaxy (MOVPE), and 3D GaN structures, InGaN quantum wells (QWs) on the 3D structures, and the doping behaviors were comprehensively studied in terms of structural, optical, and electrical properties. These basic insights led us to accomplish electrically driven polar-plane-free faceted 3D InGaN LEDs on $(\bar{1}\bar{1}2\bar{2})$. It must be noted that the radiative polychromatic emission cannot be achieved by the conventional 3D faceted LEDs with (0001) polar plane. The obtained results are summarized as follows,

In chapter 2, 3D GaN structures on $\pm(11\bar{2}2)$ GaN substrates with various mask patterns were accessed by scanning electron microscopy (SEM). The 3D shapes varied drastically depending on the surface orientations, stripe mask directions and geometries. As a remarkable result, both the $\pm(11\bar{2}2)$ surfaces with particular mask patterns provided polar-plane-free faceted 3D GaN structure with the favorable shapes for the radiative polychromatic emission properties. Then, the growth evolutions of the promising 3D structures were visualized by periodically-inserted AlGaIn marker toward tailoring the 3D shapes. It was found that the 3D GaN structures on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ exhibit distinct 3D shape developments; the 3D shape on $(\bar{1}\bar{1}2\bar{2})$ drastically changes during the growth, in contrast to the slight shape variation of the 3D shape on $(11\bar{2}2)$. As the mechanism behind it, a simple model calculation taking the inter-facet adatom migration into account suggested the drastic surface adatom migration on the 3D GaN on $(\bar{1}\bar{1}2\bar{2})$. The cause for this active migration was ascribed to the adatom kinetics on

different surface orientations. Surface morphologies of homoepitaxially-grown GaN on the relevant crystallographic orientations indicated the adatom behaviors.

In chapter 3, 3D faceted InGaN QWs grown on the polar-plane-free faceted GaN structures obtained in chapter 2 were clarified. The macroscopic and spatially resolved emission characteristics of both the 3D QWs on $(11\bar{2}2)$ and $(\bar{1}\bar{1}2\bar{2})$ were evaluated at room-temperature (RT) photoluminescence (PL) and cathodoluminescence (CL), respectively. Consequently, we found the fairly good polychromatic emission from the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ compared with the 3D QWs on $(11\bar{2}2)$, indicating the monochromatic emission and inferior QW structural property. Hereafter, this work was mainly directed to the 3D structures on $(\bar{1}\bar{1}2\bar{2})$, and the inter-facet distribution of the QW structural parameters characterized by transmission electron microscopy (TEM) were explained from the viewpoint of inter-facet adatom migration and orientation dependence of In incorporation. In addition, highly radiative polychromatic emission from the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ was demonstrated by time-resolved PL at low temperature (LT). Finally, the facet QW parameters were tuned by the growth condition, and we demonstrated pastel green and white color syntheses based on the polar-plane-free faceted 3D QWs on $(\bar{1}\bar{1}2\bar{2})$.

In chapter 4, doping condition of polar-plane-free 3D structures on $(\bar{1}\bar{1}2\bar{2})$ was examined toward the LED device operations. Here, p-type doping conditions in $(\bar{1}\bar{1}2\bar{2})$ planar LED structure were established prior to the doping of the 3D structures. The $(\bar{1}\bar{1}2\bar{2})$ planar LEDs showed quite weak electroluminescence (EL) intensities compared with the $(11\bar{2}2)$ planar LEDs grown under the same growth conditions, which was caused by the poor doping profiles revealed by secondary ion mass spectroscopy (SIMS). Accordingly, doping conditions of $(\bar{1}\bar{1}2\bar{2})$ LEDs were optimized by adjusting Mg/Ga ratios and growth pressures, and the bright EL was achieved from the $(\bar{1}\bar{1}2\bar{2})$ LEDs with improved doping profiles. The optimized doping condition for $(\bar{1}\bar{1}2\bar{2})$ LEDs was applied to the polar-plane-free 3D LED structures on $(\bar{1}\bar{1}2\bar{2})$. The SEM contrast derived from the p- and n-type conductivity enabled us to confirm the pn-junction formation on the entire 3D structure. Additionally, advanced scanning probe microscopy (SPM) techniques of scanning microwave microscopy (SMM) combined with scanning nonlinear dielectric microscopy (SNDM) gave us insight into the carrier density distribution in the cross section of the 3D LED structure and clarified the sufficient carrier densities of $\sim 10^{18} \text{ cm}^{-3}$ for the p-GaN layer and $\sim 10^{19} \text{ cm}^{-3}$ for the n-GaN layer.

In chapter 5, polar-plane-free faceted 3D InGaN-LEDs were fabricated and their device performance was characterized at RT. Device processes were optimized in advance to meet the 3D structures. Fabricated polar-plane-free LEDs on $(\bar{1}\bar{1}2\bar{2})$ exhibited

clear rectifying behaviors, and the EL spectrum was broadened from that of the $(\bar{1}\bar{1}2\bar{2})$ planar LED due to the polychromatic emission property. Then, in order to enhance the intensity of the shorter wavelength component originating from the $\{\bar{1}100\}$ side facet, 3D structures with wider side facet were introduced by using the mixed mask pattern geometry. The structure-mixed 3D LED presented the EL spectrum with enhanced shorter wavelength emission, leading to the more broadband spectrum. Finally, the structure-mixed 3D LED design was applied to synthesize white colors by increasing the In compositions. Although the EL intensity became low due to the poor material quality, the EL spectrum that shifted to the longer wavelength region fortunately synthesized the white color close to the Planckian locus.

Thus, there still remain several problems to be solved before the polar-plane-free faceted 3D LEDs reach its full potential. The author believes, however, that the present study could contribute to development of semipolar III-nitrides for highly radiative polychromatic emitters. Finally, the author would like to point out the future application of the polar-plane-free 3D LEDs. Recently, gallium nitride LED-based visible light communication is getting much attention as the new wireless communication standards [1]. High-speed InGaN-based LEDs on (0001) polar plane on sapphire substrates have already progressed to the modulation bandwidth of more than 800 MHz by the extensive researches. However, the presence of strain-induced piezoelectric fields increases the carrier lifetimes and limits further improvement of the modulation bandwidth in (0001) polar plane LEDs. In response to this challenge, nonpolar and semipolar LEDs with suppressed piezoelectric fields are regarded as the promising approaches [2]. From these situations, the polar-plane-free faceted 3D LEDs developed in this study can be expected to contribute future high-speed polychromatic emitters for visible light communication. The novel device structure might enable us to integrate tailored visible spectra synthesis and larger modulation bandwidth into one device.

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Appendix A

SEM and TEM Characterizations of 3D Faceted QWs

To evaluate inter-facet distributions of 3D faceted InGa_N quantum wells (QWs) is indispensable for understanding the origins of the polychromatic emission characteristics. X-ray diffraction (XRD) measurements are commonly used to estimate planar QW structural parameters such as In composition or thickness. However, laboratory-based macroscopic XRD measurements on 3D faceted QWs only offer limited information due to the size and the three-dimensionality. Very recently, synchrotron radiation (SR) XRD with a sub- μm spatial resolution is emerging as a technique to characterize 3D faceted QWs [1], but it is still not for general use. According to this situation, transmission electron microscopy (TEM) and energy-dispersive x-ray spectroscopy (EDS) equipped to the TEM are very powerful tools to estimate spatial distributions of the 3D faceted QWs. This study also conducted TEM + EDS to assess local QW structural property of the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$. However, TEM measurements require complicated sample preparations and are very time-consuming; the TEM specimen must be thin (about 100 nm or less) to transmit sufficient electrons to form an image, and the specimen preparation procedures such as Ar ion milling should be performed. This simultaneously means that the sample is cracked and broken through the process.

On the other hand, scanning electron microscopy (SEM) and EDS equipped to the SEM do not require a complicated specimen preparation and are commonly used to characterize micro-scale structures, though the spatial resolution is larger than TEM due to the spreading of incident primary electrons. In other words, SEM + EDS can evaluate the QW structure correctly if the sample dimension is larger than the spatial resolution. In the following, inter-facet distributions of 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ estimated

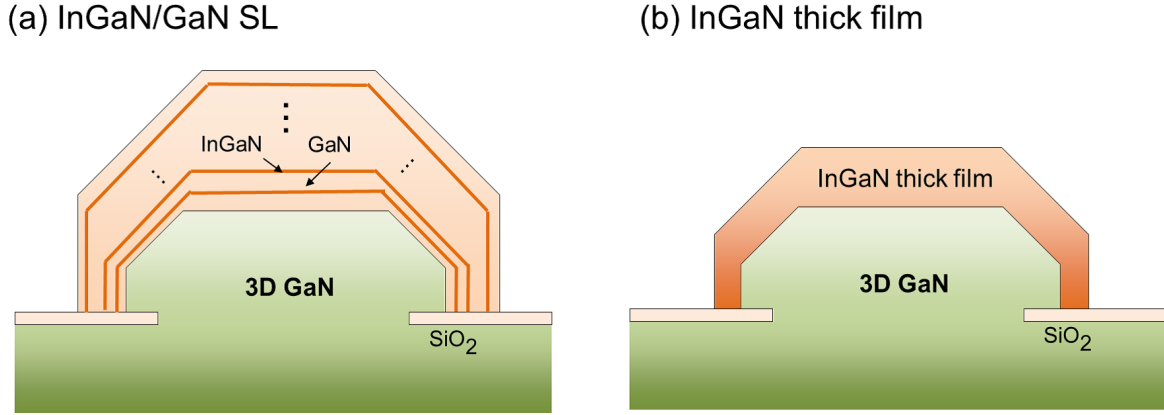


Figure A.1: Schematic illustrations of (a) InGaN/GaN SL and (b) InGaN thick film on 3D GaN structures.

by TEM + EDS and SEM + EDS are compared, and it is demonstrated that both the measurements provide essentially the same results.

In order to estimate local InGaN thickness and In composition of 3D QWs by SEM + EDS, thick InGaN layers or 200-period InGaN/GaN superlattices (SLs) were grown on 3D GaN structures instead of MQWs for the thickness or composition estimations, respectively. QWs are not suitable because of the experimental resolution larger than the dimensions of QWs. Figure A.1 illustrates schematics of (a) InGaN SL and (b) InGaN thick film on 3D GaN structures. The total thickness of SLs was set to be more than the resolution of EDS (about 100 nm). The growth conditions such as substrate temperature, pressure, and V/III ratio are, of course, the same as the target 3D QWs. The local InGaN thickness is estimated from the cross-sectional SEM image of InGaN thick film. Meanwhile, the local In composition is estimated from the cross-sectional EDS of InGaN/GaN SL. However, as a matter of course, In compositions estimated from the InGaN/GaN SL correspond to the effective In compositions of InGaN/GaN SL, not those of the InGaN layers. Thus, the effective In compositions are calibrated to the In compositions of the InGaN layers using the thickness and growth times of the InGaN thick film and InGaN/GaN SL. The In composition at InGaN well, x_{well} , can be expressed by

$$x_{\text{well}} = \frac{d_{\text{SL}}}{d_{\text{InGaN-thick}} \times \frac{t_{\text{SL}}}{t_{\text{InGaN-thick}}}} \times x_{\text{ave.}}, \quad (\text{A.1})$$

where d_{SL} and $d_{\text{InGaN-thick}}$ are local thicknesses of the InGaN SL and InGaN thick film, t_{SL} and $t_{\text{InGaN-thick}}$ are total growth times of the InGaN layer within the InGaN SL and

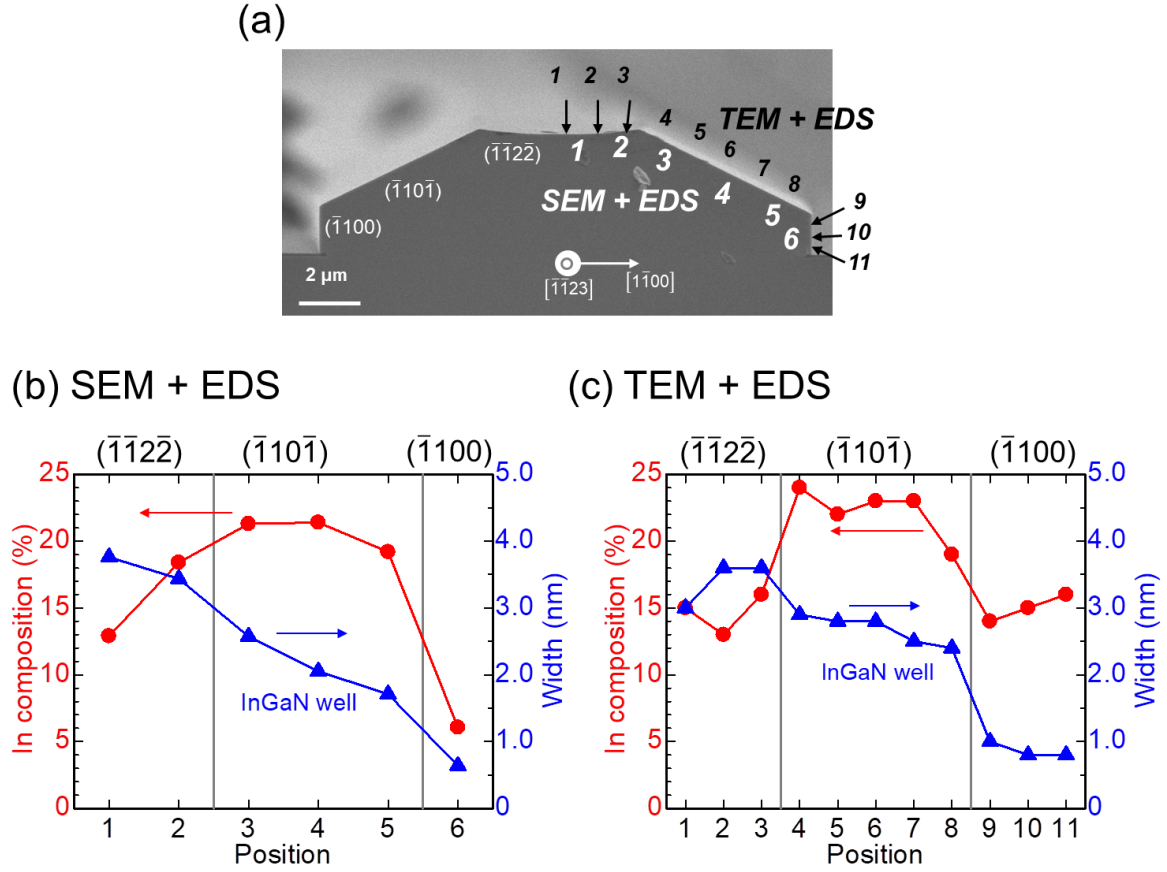


Figure A.2: Local In compositions and InGaN well widths estimated by (a) SEM + EDS and (b) TEM + EDS measurements.

the InGaN thick film, $x_{\text{ave.}}$ is a measured In composition of the InGaN SL. It should be noted that InGaN thick film is not suitable for In composition evaluation, because the InGaN thickness is usually beyond the critical thickness of lattice relaxation, and the relaxed InGaN layers incorporated more In atoms than coherently-grown InGaN layers. Thus, the In composition estimated from the relaxed InGaN layers should be higher than that of the 3D QWs.

Figure A.2 compares local In compositions and InGaN well thicknesses of (a) polar-plane-free faceted 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ estimated by (b) SEM + EDS and (c) TEM + EDS. For SEM results, InGaN well thickness varies in the order of $(\bar{1}\bar{1}2\bar{2}) > (\bar{1}\bar{1}0\bar{1}) > (\bar{1}\bar{1}00)$, and In compositions are $(\bar{1}\bar{1}0\bar{1}) > (\bar{1}\bar{1}2\bar{2}) > (\bar{1}\bar{1}00)$. Comparing with the TEM results [Fig. A.2(b)], both the variations of In compositions and InGaN well thickness are quite similar to each other. Although the In composition at $(\bar{1}\bar{1}00)$ facet by SEM is smaller than that by TEM, this would be due to the small EDS signal of the thinner

InGaN/GaN SL thickness. From these results, both SEM + EDS and TEM + EDS are considered to provide essentially the same results. The QW structural characterization using SEM + EDS was also applied for the 3D QWs on (0001), and the validity was confirmed [2, 3].

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Appendix B

$(\bar{1}\bar{1}2\bar{2})$ planar InGaN QWs

Here, let us discuss interface abruptness of $(\bar{1}\bar{1}2\bar{2})$ planar quantum wells (QWs) and the potential of $(\bar{1}\bar{1}2\bar{2})$ orientation for future light-emitting devices.

Firstly, regarding the QW structures, Nishinaka *et al.* previously reported that $(\bar{1}\bar{1}2\bar{2})$ QWs exhibit rough interfaces in contrast to smooth interfaces of $(11\bar{2}2)$ QWs grown under the same growth conditions [1], as shown in Fig. B.1(a). They also found that $(\bar{1}\bar{1}2\bar{2})$ GaN homoepitaxial layers grown under H_2 carrier gas were atomically flat in a wide range of growth temperature and V/III ratio than $(11\bar{2}2)$ GaN homoepitaxial layers, thus the reason of the poor abruptness of the $(\bar{1}\bar{1}2\bar{2})$ QWs was attributed to the N_2 carrier gas during the QW growth. A part of the reason why Nishinaka selected $(11\bar{2}2)$ surface as the substrate orientation for $\{11\bar{2}2\}$ laser diodes (LDs) was the superior QW structure [2]. However, during the investigations of 3D QWs on $(\bar{1}\bar{1}2\bar{2})$ (Chap. 3), we found relatively smooth interfaces of $(\bar{1}\bar{1}2\bar{2})$ facet QWs presented in Fig. B.1(b). It should be noted that the carrier gas used in QW growth was N_2 , the same as the previous study in Fig. B.1(a). This finding motivates us to investigate the reason. To fairly discuss the previous study and this observation, planar QWs grown under the same growth conditions as the 3D QWs were also observed, and the result is shown in Fig. B.1(c). Since inter-facet adatom migration from the inclined $\{\bar{1}10\bar{1}\}$ to top $(\bar{1}\bar{1}2\bar{2})$ facets occurs during the growth of the 3D QWs, the situation of growth dynamics is considered to be different from the planar QWs, as discussed in Sec. 3.5. However, the planar QWs in this study also exhibit smooth interface abruptness compared with the previous study. This suggests that the inter-facet adatom migration of the 3D QWs is not a dominant factor to improve the interface abruptness, and also indicates the minor role of N_2 carrier gas.

The reason is under investigation, but one of the plausible factors is the InGaN

growth rates. In the previous study [Fig. B.1(a)], the InGaN growth rate was 0.17 nm/s, whereas those of the facet and planar QWs in this study [Fig. B.1(b) and B.1(c)] is 0.11 and 0.08 nm/s, respectively. The slower growth rate usually promotes surface migration of adatoms, resulting in the smooth surface. Additionally, the TEM image of the previous study indicates that the interface abruptness becomes worse by inserting InGaN well layers; the final GaN barrier layer at the surface side is more degraded than the first GaN barrier layer. This observation also suggests the crucial role of the smooth InGaN surface for achieving the superior interface abruptness.

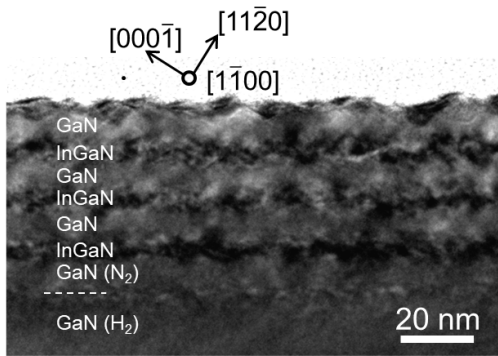
The abrupt interface was still observed in the case of higher growth pressure (550 Torr), which was employed in chapters 4 and 5. We confirmed almost no growth pressure dependence of growth rates, and the InGaN growth rate was 0.10 nm/s. This result is consistent with the above consideration.

Finally, let us describe the superior characteristics of $(\bar{1}\bar{1}2\bar{2})$ orientation for future light emitters. As mentioned above, Nishinaka *et al.* previously selected $(11\bar{2}2)$ orientation as the substrate for LD structures partially due to the interface abruptness of the QW structure. However, this study found out the improved QW structure on $(\bar{1}\bar{1}2\bar{2})$ surface comparable to that on $(11\bar{2}2)$ surface. Thus, at present, both orientations are considered to be promising. In addition, from the viewpoint of impurity incorporation, $(11\bar{2}2)$ surface exhibits numerous incorporation of oxygen (O) impurity even under higher growth temperatures. On the other hand, O concentrations [O] in epitaxial layers can be significantly suppressed by the use of $(\bar{1}\bar{1}2\bar{2})$ surface, as demonstrated in chapter 4. This characteristic plays a crucial role in fabricating LD structures. A typical LD structure is illustrated in Fig. B.2. As a matter of course, unintentional impurities often degrade device performances, and in particular, surface orientations with numerous O incorporation behaviors cause difficulties in fabricating the p-InGaN guide layer with satisfactory electrical characteristics. This is because, lower growth temperatures are usually applied to InGaN layers and simultaneously increase [O], which is well-known as a compensator for acceptors. Consequently, the p-InGaN guide layer tends to be resistive, degrading the current-voltage characteristics of the LD operations. On the other hand, $(\bar{1}\bar{1}2\bar{2})$ with extremely suppressed O incorporation behavior can be expected to show the superior device performance. Therefore, the $(\bar{1}\bar{1}2\bar{2})$ plane is quite attractive for highly efficient semipolar light emitters in future optoelectronics.

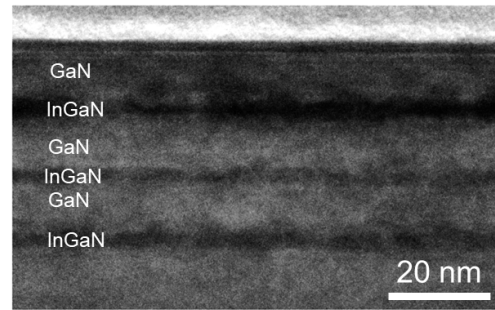
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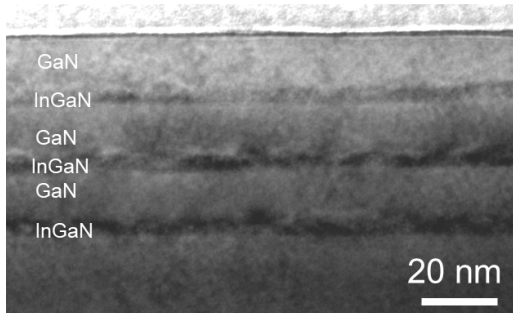
(a) Previous study (300 Torr)



(b) Facet QWs (300 Torr)



(c) Planar QWs (300 Torr)



(d) Planar QWs (550 Torr)

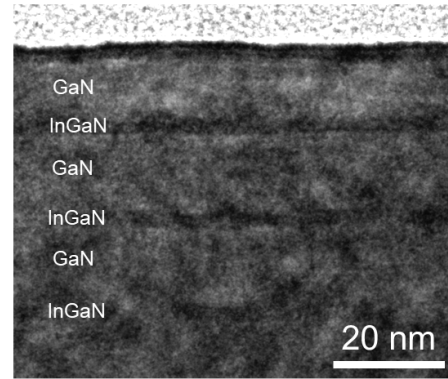


Figure B.1: TEM images of $(\bar{1}\bar{1}2\bar{2})$ QWs. (a) The previous report [1]. (b) The top $(\bar{1}\bar{1}2\bar{2})$ facet QW of the 3D QWs on $(\bar{1}\bar{1}2\bar{2})$, and planar QWs grown at (c) 300 Torr and (d) 550 Torr fabricated in this study.

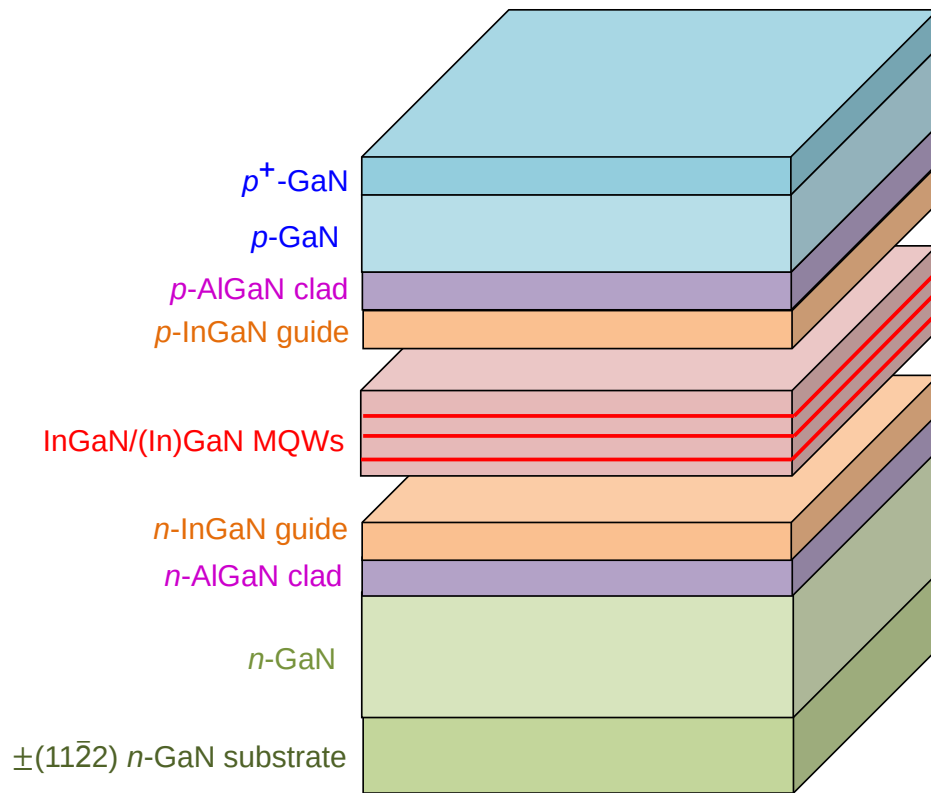


Figure B.2: Schematic of a LD structure on $\pm(11\bar{2}2)$ GaN substrate.

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Appendix C

Remaining Issues for Polar-Plane-Free Faceted 3D LEDs

In this appendix, let us explain remaining issues for polar-plane-free 3D LEDs on $(\bar{1}\bar{1}2\bar{2})$. The clear rectifying property and broad electroluminescence (EL) spectra were successfully achieved, but the device performance is still far from ideal operations.

One of the issues is insufficient current spreading in the 3D LEDs. Figures C.1(a) and C.1(b) show photographs of the 3D LED chip and the emission operated under direct current (DC) of 20 mA, respectively. Their locations are the same as each other. Figure C.1(b) clearly shows only a part of the LED chip within the size of $350 \mu\text{m}^2$ showing EL, and the other part looks dark. This observation indicates the poor electrical injection into the entire chip area. Since the n-electrode was formed entirely on the sample backside, the attention should be directed to the p-electrode.

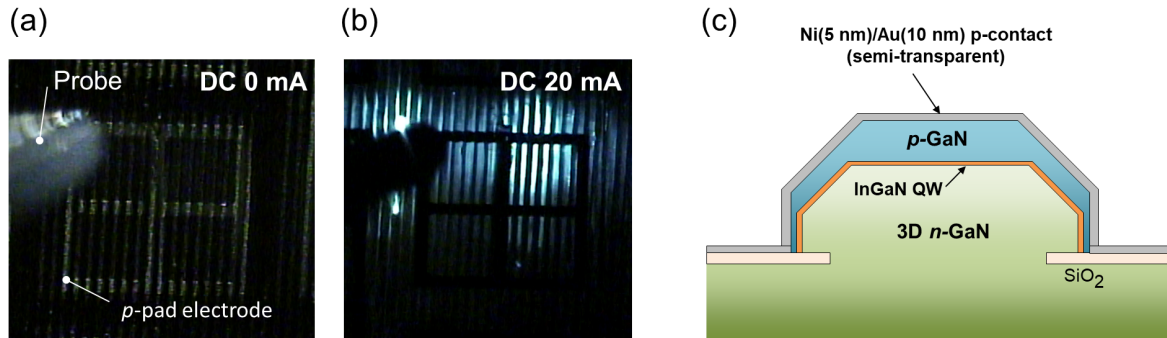


Figure C.1: Example photographs of a LED chip under (a) no operation and (b) DC 20 mA at RT. (c) Schematic of a 3D LED structure on $(\bar{1}\bar{1}2\bar{2})$ with a Ni/Au p-contact electrode.

Moreover, we confirmed the conductivity of the p-pad electrode comparable to the p-pad electrode of a $(\bar{1}\bar{1}2\bar{2})$ planar LED. Thus, the nonuniform EL could be attributed to the p-contact electrode. The severe thickness design of Ni (5 nm)/Au (10 nm) p-contact must be preserved in order to form ohmic contact to p-GaN together with the electrode transparency. Although we attempted the uniform p-contact formation on entire 3D structures by the oblique-angle vacuum evaporation (Sec. 5.2), it should include the thickness distribution among the facets. The situation is illustrated in Fig. C.1(c). Such a distribution may disturb the efficient current spreading over the whole chip area.

Indium-Tin-Oxide (ITO) p-contact is an attractive approach to solve this issue because ITO film has high transparency in the visible spectral region and allows us to form ohmic contact to p-GaN [1–4]. Thus, severe thickness design is unnecessary for the ITO p-contact. ITO p-contact has already been implemented in commercially available InGaN LEDs [3]. Another advantage is that ITO films can be deposited by a sputtering technique. The relatively higher gas pressures and larger target dimensions are beneficial for covering entire 3D structures without elaborate preparation.

Another issue for the device operations of the polar-plane-free 3D LEDs on $(\bar{1}\bar{1}2\bar{2})$ is leakage current. Although clear rectifying property appears under appropriate diode operations, a serious leakage current is also frequently observed in some cases, resulting in no rectifying property, as shown in Fig. C.2(a). It is difficult to identify the leakage current pass, but the emission photograph in Fig. C.2(b) indicates a clue; spot-like violet emission can be observed under the operation. Looking back at the EL spectra of the 3D LEDs on $(\bar{1}\bar{1}2\bar{2})$ in Fig. 5.6, violet emission component relies on the $\{\bar{1}100\}$ side facet. Therefore, the spot-like violet emission could be attributed to the QW emission from the $\{\bar{1}100\}$ side facet, suggesting the leakage current path through the $\{\bar{1}100\}$ facet QWs as shown in Fig. C.2(c).

If this is the case, efficient p-type doping on the side facet is considered to be hindered by for some reason. One of the possible reasons is a slow growth rate of the $\{\bar{1}100\}$ facet, as indicated in Fig. 4.15. A part of the p-GaN layer on the $\{\bar{1}100\}$ facets may be too thin to form the proper pn-junction. The second reason is impurity incorporation behavior of $\{\bar{1}100\}$ orientation, which was reported to have relatively higher incorporation of O by Cruz *et al.* [5]. The $\{\bar{1}100\}$ facet may have poor p-type conductivity, leading to the leaky characteristic. The final possible reason is O and Si impurity diffusions from the SiO₂ mask. Matthews *et al.* performed spatially resolved Raman spectroscopy of lateral epitaxially overgrown GaN on (0001) GaN template and found that carrier concentrations are extremely high ($\sim 10^{20}$ cm⁻³) above SiO_x mask

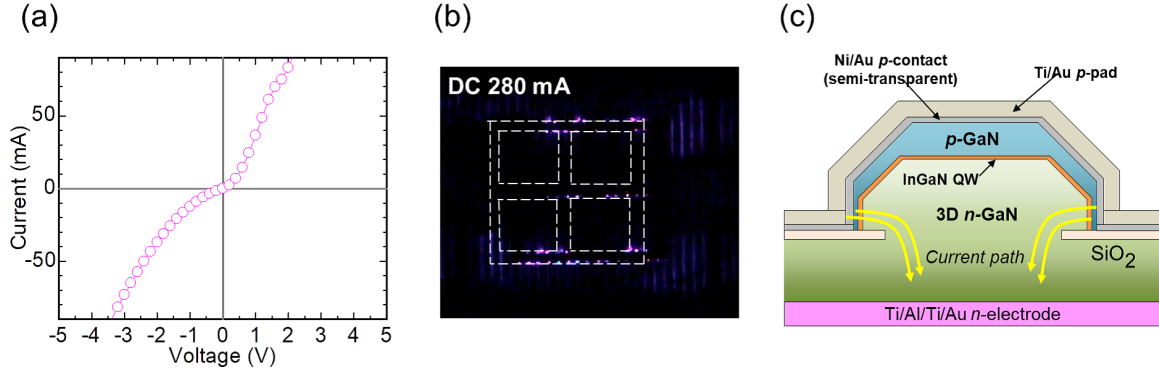


Figure C.2: (a) Current-voltage characteristic and (b) an example photograph under DC 280 mA of the polar-plane-free 3D LED on $(\bar{1}\bar{1}2\bar{2})$ with leaky diode property. (c) Schematic diagram of leakage current path presumed from the emission in (b).

layers probably due to the dopant diffusion from the SiO_x mask [6]. In our 3D structures, the $\{\bar{1}100\}$ facet is above SiO₂ mask layer, and the dopant diffusion can prevent efficient p-type doping. Recently, Rishinaramangalam *et al.* proposed that n-AlGaIn underlayer can enhance Mg incorporation and lower Si and O impurity concentrations within the p-GaN layer of 3D InGaIn LED structures on (0001) GaN templates [7]. In any case, we need further improvement of the doping conditions or structure designs for efficient p-type doping.

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List of Publications

A. Full Length Papers and Letters

1. Y. Matsuda, M. Funato, and Y. Kawakami,
“Realization of multi-wavelength emission using polar-plane-free InGaN multi-facet quantum wells,”
IEICE Technical Report **116**, 67 (2016).
2. Y. Matsuda, M. Funato, and Y. Kawakami,
“Polar-plane-free faceted InGaN microstructures for highly efficient polychromatic emitters,”
IEICE Technical Report **117**, 5 (2017).
3. Y. Matsuda, M. Funato, and Y. Kawakami,
“Polychromatic emission from polar-plane-free faceted InGaN quantum wells with high radiative recombination probabilities,”
Appl. Phys. Express **10**, 071003 (2017).
4. Y. Matsuda, M. Funato, and Y. Kawakami, **(Special Section on Distinguished Papers)**
“Growth mechanism of polar-plane-free faceted InGaN quantum wells,”
IEICE TRANS. ELECTRON. **E101-C**, 532 (2018).

B. International Conferences

1. Y. Matsuda, M. Funato, and Y. Kawakami, **(Landmark Lecture)**
“InGaN-based three-dimensional structures for phosphor-free multi-wavelength emitters,”
15th International Symposium on the Science and Technology of Lighting, Kyoto University, Japan, 24P-LL2, (May. 2016), oral and poster.

2. Y. Matsuda, M. Funato, and Y. Kawakami,
 “Polychromatic emission from semi/nonpolar faceted 3D-InGaN quantum wells with high radiative recombination probabilities,”
12th International Conference on Nitride Semiconductors, Strasbourg, France, B1.55, (July. 2017), poster.
3. Y. Matsuda, M. Funato, and Y. Kawakami,
 “Growth evolution of polar-plane-free faceted GaN structures,”
19th International Conference on Metalorganic Vapor Phase Epitaxy, Nara, Japan, 4C-2.5, (June. 2018), oral.
4. Y. Kawakami, Y. Matsuda, T. Ozaki, K. Kataoka, and M. Funato, **(Plenary)**
 “Nitride-based 3D-structures for polychromatic LEDs,”
16th International Symposium on the Science and Technology of Lighting, The University of Sheffield, UK, (June. 2018), oral.
5. Y. Matsuda, M. Funato, and Y. Kawakami,
 “Polar-plane-free faceted InGaN quantum wells toward highly radiative pastel and white color syntheses,”
International Workshop on Nitride Semiconductors 2018, Kanazawa, Japan, GR13-1, (November 2018), oral.
6. Y. Matsuda, M. Funato, and Y. Kawakami,
 “Exploring the growth procedures for polar-plane-free faceted InGaN-LED structures,”
The 7th International Conference on Light-Emitting Devices and Their Industrial Applications, Yokohama, Japan, LEDIA-7-04, (April. 2019), oral.
7. Y. Matsuda, M. Funato, and Y. Kawakami,
 “Fabrication of polar-plane-free faceted InGaN LED structures with polychromatic emission properties,”
13th International Conference on Nitride Semiconductors, Washington, USA, A12-05, (July 2019), oral.
8. Y. Matsuda, M. Funato, and Y. Kawakami,
 “Experimental Study on Semipolar ($11\bar{2}2$) LEDs toward Polar-Plane-Free Faceted InGaN LEDs on ($11\bar{2}2$),”
Asia-Pacific Workshop on Widegap Semiconductors 2019, Okinawa, Japan, ThP-OD-13, (Nov. 2019), poster.

C. Domestic Conferences

1. Y. Matsuda, M. Funato, and Y. Kawakami,
 “Three dimensional semi/nonpolar InGa_N quantum wells toward phosphor-free polychromatic emitters,”
35th Electronic Materials Symposium, Th2-3, Laforet Biwako, (Jul. 2016), short presentation and poster.
2. Y. Matsuda, M. Funato, and Y. Kawakami,
 “Realization of multi-wavelength emission using polar plane-free InGa_N multi-facet quantum wells,”
The Institute of Electronics, Information and Communication Engineers, ED2016-70, Kyoto Univ., (Dec. 2016), oral.
3. Y. Matsuda, M. Funato, and Y. Kawakami,
 “Growth and characterization of polar plane-free 3D faceted InGa_N quantum wells,”
64th Japan Society of Applied Physics Spring Meeting, 15a-503-11, Pacifico Yokohama, (Mar. 2017), oral.
4. Y. Matsuda, M. Funato, and Y. Kawakami,
 “Consideration of inter-facet variations of In composition in 3D faceted InGa_N quantum wells,”
78th Japan Society of Applied Physics Fall Meeting, 8a-A301-6, Fukuoka Convention Center, (Sep. 2017), oral.
5. Y. Matsuda, M. Funato, and Y. Kawakami,
 “Growth mechanism of polar-plane-free faceted InGa_N quantum wells with high radiative recombination probabilities,”
36th Electronic Materials Symposium, Fr1-10, Laforet Biwako, (Nov. 2017), short presentation and poster.
6. Y. Matsuda, M. Funato, and Y. Kawakami, **(Invited)**
 “Polar-plane-free faceted InGa_N microstructures for highly efficient polychromatic emitters,”
The Institute of Electronics, Information and Communication Engineers, LQE2017-86, Kikai Shinko Kaikan building, (Dec. 2017), oral.

7. Y. Matsuda, M. Funato, and Y. Kawakami,
“Pastel color syntheses and control based on polar-plane-free faceted 3D InGaN quantum wells,” **(Young Scientist Presentation Award)**
Nanocrystalline Materials and Epitaxial Crystal Growth 2018, Th-P19, Nagoya Univ., (Jul. 2018), short presentation and poster.
8. Y. Matsuda, M. Funato, and Y. Kawakami,
“Pastel and white color syntheses using polar-plane-free faceted InGaN quantum wells,”
79th Japan Society of Applied Physics Fall Meeting, 20a-146-12, Nagoya Congress Center, (Sep. 2018), oral.
9. Y. Matsuda, M. Funato, and Y. Kawakami,
“Pastel green synthesis from polar-plane-free faceted 3D InGaN quantum wells with high radiative recombination probabilities,” **(Young Scientist Presentation Award)**
1st Crystal Engineering and ISYSE Joint Meeting, 2P025, Kogakuin Univ., (Nov. 2018), short presentation and poster.
10. Y. Matsuda, M. Funato, and Y. Kawakami,
“Crystal growth and evaluation of polar-plane-free faceted InGaN LED structure,”
66th Japan Society of Applied Physics Spring Meeting, 9a-W541-1, Tokyo Tech., (Mar. 2019), oral.
11. Y. Matsuda, M. Funato, and Y. Kawakami,
“Experimental study on device processes of polar-plane-free faceted InGaN LEDs,”
80th Japan Society of Applied Physics Fall Meeting, 19a-E310-7, Hokkaido Univ., (Sep. 2019), oral.
12. Y. Matsuda, M. Funato, and Y. Kawakami,
“Polar-Plane-Free Faceted InGaN-LEDs toward Next-Generation White Light Sources,” **(Best Poster Presentation Award)**
The 2nd Nanomics Symposium, 3, Kyoto Univ., (Oct. 2019), short presentation and poster.

13. Y. Matsuda, M. Funato, and Y. Kawakami,
 “Electroluminescence from Polar-Plane-Free Faceted InGaN LEDs,”
67th Japan Society of Applied Physics Spring Meeting, 13a-A302-2, Sophia Univ.,
 (Mar. 2019), oral.

D. Awards

1. IEICE Electronics Society LQE Young Researchers Award
 (IEICE Electronics Society LQE meeting 2016)
2. Best Poster Award
 (International Symposium on Photonics and Electronics Science and Engineering
 2018)
3. Young Scientist Presentation Award
 (Nanocrystalline Materials and Epitaxial Crystal Growth 2018)
4. Young Scientist Presentation Award
 (1st Joint Meeting of CST and ISYSE)
5. Student’s Paper Award “Yamaguchi Masahito Award”
 (The 7th International Conference on Light-Emitting Devices and Their Industrial
 Applications)
6. Best Poster Presentation Award
 (The 2nd Nanomics Symposium)

E. Research Grants

1. Grant-in-Aid for JSPS Research Fellow (DC2)
2. Innovation of Advanced Photonic and Electronic Devices Grant-in-Aid
3. ICOM Foundation
4. The Kyoto University Foundation