SPECIAL ISSUE: Optical Gain Materials towards Enhanced Light-Matter Interactions

Design and growth of GaN-based blue and green laser diodes

Aiqin Tian^{1†}, Lei Hu^{1,2†}, Liquan Zhang¹, Jianping Liu^{1,2*} and Hui Yang^{1,2}

ABSTRACT GaN-based laser diodes (LDs) extend the wavelength of semiconductor LDs into the visible and ultraviolet spectrum ranges, and are therefore expected to be widely used in quantum technology, bio & medical instruments, laser displays, lighting and materials processing. The development of blue and green LDs is still challenging, even though they are based on the same III-nitride materials as GaN-based light-emitting diodes. The challenges and progress of GaN-based blue and green LDs are reviewed from the aspects of epitaxial growth and layer structure design. Due to large differences in lattice constants and growth conditions for InN, GaN, and AlN, considerable effort is required to improve the quality of InGaN multiple quantum well (MQW) gain medium for blue and especially green LDs. p-type doping profiles, conditions and layer structures are critical to reduce the internal losses and to mitigate the degradation of InGaN MQWs. Hole injection is also a key issue for GaN-based LDs.

Keywords: GaN-based LDs, InGaN, thermal degradation, In segregation, optical loss, carrier injection

INTRODUCTION

The invention and development of GaN-based light-emitting diodes (LEDs) have extended the wavelengths of semiconductor optoelectronic devices into the visible and ultraviolet spectrum ranges. GaN-based blue LEDs have been widely used in lighting and displays, and the Nobel Prize in Physics of 2014 honored the inventors of efficient blue LEDs—Professors Akasaki I, Amano H and Nakamura S [1–5]. GaN-based laser diodes (LDs), however, are different kinds of light sources with high brightness (4 orders of magnitude brighter than that of LEDs) and high spectral purity. Semiconductor LDs are now the

most widely used lasers because of low costs, small sizes, high efficiencies and long lifetimes. Like the LEDs, GaN-based LDs have extended the visible and ultraviolet spectrum ranges. GaN-based violet LD with wavelength of 405 nm for the application of high-density optical storage was the first research focus of GaN-based LDs. GaN-based blue and green LDs have been studied intensely due to great potential for wide applications in displays, lighting, quantum technology, optical clock, medical instruments, materials processing and underwater communications and detection (Fig. 1a).

Laser displays using red, green and blue LDs are emerging technologies with larger color gamuts, higher color saturation, and capabilities for both pico-projectors and larger displays [6–11]. Fig. 1b compares the color gamuts of laser displays vs. other technologies [11]. GaN-based direct blue and green emission semiconductor LDs are thus desirable for laser displays. However, their fabrication is more challenging, compared with that of LEDs [12,13].

There are four main difficulties to fabricate high-performance GaN-based blue and green LDs. First, as shown in Fig. 2, the LD epitaxial structures are more complex and more strained to form the optical cavity, which increases the complexity of epitaxial growth and causes more crystalline defects. Second, LDs are more sensitive to defects, including non-radiative recombination centers and inhomogeneity, both of which reduce the peak gain of LDs. Meanwhile, InGaN-based LEDs are not sensitive to defects because of the localization effect. Spectral broadening due to indium (In) composition fluctuations and interface roughness is a particular issue for green LDs whose gain medium is high-In-content InGaN multiple

¹ Key Lab of Nanodevices and Applications, Suzhou Institute of Nano-Tech and Nano-Bionics, Chinese Academy of Sciences, Suzhou 215123, China

² School of Nano-Tech and Nano-Bionics, University of Science and Technology of China, Hefei 230026, China

[†] These two authors contributed equally to this work.

* Corresponding author (email: jpliu2010@sinano.ac.cn)

Figure 1 (a) Applications of GaN-based blue and green LEDs. (b) Color gamut in the Commission Internationale de l'Eclairage (CIE) chromaticity diagram of laser, LED, liquid crystal display (LCD) and cathode ray tube (CRT) display [11].

Figure 2 Schematic of a conventional GaN-based LED with a palladium (Pd)/platinum (Pt)/gold (Au) electrode or a hybrid GaN-based LED with indium tin oxide (ITO) cladding layer. Reprinted with permission from Ref. [29]. Copyright 2020, Chinese Laser Press.

quantum wells (MQWs) [14]. Third, LEDs are more sensitive to impurities because light will be strongly absorbed when propagating forward and backward many times within the cavity. Fourth, hole injection is usually inhomogeneous among MQWs because of the large effective masses of holes and the large potential barrier. Lack of holes in n-side quantum wells (QWs) will lead to high light absorption and, therefore, high threshold currents and reduced slope efficiencies for the LEDs. In LEDs, either holes are injected into bottom QWs *via* V-pit sidewalls, or without hole injection, and light emission in the bottom QWs is not an issue.

The first GaN-based blue LEDs were invented in 1999 by Nichia [15], while the first high-brightness LEDs was reported in 1993. The performance was improved in 2001 by using a free-standing GaN substrate [16]. The output power of GaN-based blue LEDs was improved to 200 mW in 2005 [17], 500 mW in 2006 [18], 1 W in 2008 [19], and 5.25 W most recently [20]. However, the epitaxial growth

and layer structures of GaN-based LEDs have not been reported in significant detail. We have also obtained blue LEDs with output powers more than 2 W [21], and recently have improved slope efficiencies to 1.65 W A^{-1} [21,22]. The development of GaN-based green LEDs ($\lambda > 500 \text{ nm}$) has been even more challenging. They were reported in 2009 by Osram Corp. [6]. Since then, green InGaIn LEDs grown on *c*-plane [6,23–29], (11 $\bar{2}$ 2) plane [30], and (20 $\bar{2}$ 1) plane [31–34] have been realized. Challenges and recent progresses on GaN-based blue and green LEDs will be discussed below.

CHALLENGES

Crystalline defects

The first challenge is the preparation of high-quality InGaIn/GaN MQWs, which is the gain medium of LEDs, especially for high-In-content InGaIn/GaN MQW green LEDs. The quantum efficiency of GaN-based LEDs drops significantly as emission wavelength extends into the “green gap” [35,36], as shown in Fig. 3a. The efficiency of GaN-based LEDs drops even more severely due to more complex structures. Table 1 lists the properties and general growth conditions of GaN, AlN and InN [37]. The very different lattice constants and bond strengths result in large strains in the LED structures, and very different growth conditions are needed for different layers. Approximately 30% In must be incorporated in InGaIn QWs to realize green LEDs, which results in crystalline defects in three ways. First, because of weak In–N bonds, the growth temperature for InGaIn must be lowered to incorporate sufficient In. Consequently, both the atomic diffusion length and the NH_3 decomposition efficiency decrease at low temperatures, which results in crystalline

Figure 3 (a) External quantum efficiency vs. peak wavelength for high power LEDs based on III-nitride and III-phosphide systems. Reprinted with permission from Ref. [36]. Copyright 2008, John Wiley & Sons. (b) Comparison of measured modal peak gain vs. current for 450 and 490 nm LDs. Reprinted with permission from Ref. [39]. Copyright 2011, AIP Publishing LLC.

defects, fluctuations of In composition, and rough interfaces in InGa_N QWs because the growth temperature must be high enough to maintain a step-flow growth which is essential for a high-quality epitaxial growth [38]. Inhomogeneous broadening caused by In composition fluctuations and rough interfaces in InGa_N QWs significantly lower the modal peak gain of green LDs [39], as shown in Fig. 3b. Secondly, the ideal growth temperature for a p-AlGa_N cladding layer is as high as 1000°C to ensure low resistance. However, such high temperature will induce serious thermal degradation for both blue and green InGa_N QWs [16,40–44]. Thirdly, the lattice mismatch between InGa_N QWs and Ga_N is as large as 1.6% for blue QWs and 3.3% for green QWs, which induces crystalline defects in InGa_N QWs. Crystalline defects not only reduce the radiative efficiency [45], but also shorten the device lifetimes [46].

Light absorption

The ionization energy of a magnesium (Mg) dopant in p-Ga_N is as large as 160 meV, and increases with aluminum (Al)-content in p-AlGa_N [47,48], making it the best dopant for p-(Al)Ga_N. Therefore, only less than 10% of Mg atoms are ionized and become free holes, and more than 90% form acceptor-bound holes, which are the main origin of light absorption in Ga_N-based LDs and create large internal optical losses [49–52]. Large internal optical losses lead to high threshold currents and low slope efficiencies [21,49].

Carrier injection

Hole injection is usually inhomogeneous among MQWs because of the large effective masses (m_0) of holes and potential barrier, as reported extensively [53–55]. The effective masses of heavy holes in Ga_N and In_N approach

Table 1 Properties and general growth conditions of Ga_N, Al_N and In_N [37]

Sample	In _N	Ga _N	Al _N
Lattice mismatch (%)	11	0	−2.4
Bond strength (eV)	1.93	2.20	2.88
V/III ratio	30,000	2000	200
Growth temperature (°C)	500–600	1000–1100	1350–1450

1.96 and 1.67 m_0 , respectively [56]. Moreover, the tilt of the potential barrier caused by the polarization field increases the hole transport barrier and makes n-side hole injection into the QWs difficult [57]. The lack of holes in the n-side QWs results in high light absorption, and therefore high threshold current and reduced slope efficiency for LDs [58,59].

Quantum confined Stark effect

Ga_N-based materials with *c*-plane orientations are the most mature for epitaxial growth. However, the quantum confined Stark effect (QCSE) is a challenge for fabricating *c*-plane green LDs. There are many detailed reports [60–65] about QCSE, which originates from the polarization electric fields caused by both spontaneous and piezoelectric polarization charges at the heterointerface [66,67]. The polarization electric field has been estimated to be more than 2 MV cm^{−1} in green InGa_N QWs [68,69]. Consequently, the overlap of electron-hole (e-h) wavefunctions is greatly reduced, as shown in Fig. 4, which results in low luminescence efficiency.

Because *c*-plane InGa_N QWs suffer from the QCSE, nonpolar (*a*-plane, *m*-plane) InGa_N QWs free from QCSE, and semipolar (*r*-plane) InGa_N QWs with small QCSE, have advantages for fabricating high performance green LDs [70]. Some groups (Sony & Sumitomo [34,71],

Figure 4 Schematic band diagram, and electron and hole wavefunctions without (a) and with (b) QCSE.

University of California at Santa Barbara (UCSB) [72,73] have demonstrated green LDs based on these planes. However, crystalline defects such as stacking faults [74] and misfit dislocations readily appear in nonpolar and semipolar InGaN QWs, which limit the utilization of those planes.

PROGRESS OF BLUE LDs

Epitaxial growth: suppression of dark spots

Blue LD epitaxial structures were grown *via* a metal-organic chemical vapor deposition (MOCVD) on *c*-plane GaN substrates with a dislocation density of 10^6 cm^{-2} or lower and then fabricated into ridge waveguide LD chips, schematically shown in Fig. 2. The epitaxial structure consisted of a silicon (Si)-doped $\text{n-Al}_{0.08}\text{Ga}_{0.92}\text{N}$ cladding layer, a Si-doped n-GaN layer, a Si-doped $\text{n-In}_{0.04}\text{Ga}_{0.96}\text{N}$ waveguide layer, two-period unintentionally-doped MQWs, an unintentionally-doped $\text{In}_{0.02}\text{Ga}_{0.98}\text{N}$ waveguide layer, a Mg-doped $\text{p-Al}_{0.2}\text{Ga}_{0.8}\text{N}$ electron blocking layer, a 500 nm Mg-doped p-AlGaIn cladding layer with 7% average Al-content (p-AlGaIn/p-GaN superlattice structures with periodic 2.5 nm p-AlGaIn and 2.5 nm p-GaN to reduce resistivity), a p-InGaIn contact layer, and a Pd/Pt/Au metal p-electrode .

The growth temperature for blue InGaN QWs is around 700°C , while it is $950\text{--}1000^\circ\text{C}$ for a p-AlGaIn cladding layer. During the high-temperature epitaxial growth of the 500 nm p-AlGaIn cladding layer, a large thermal budget was imposed on the InGaN QWs, which thermally decompose. A Nikon A1 inverted laser confocal microscope with a 405 nm LD excitation laser source was used to image the luminescence of the blue QW active region of LD structures. Thermal decomposition of InGaN QWs leads to the formation of non-radiative recombination centers, which appear as dark spots in the image [16]. Fig. 5a–c show the fluorescence images of blue LD wafers with p-AlGaIn cladding layers grown at temperatures of 960, 950 and 940°C , respectively. The density of dark spots significantly decreased with the growth temperature of the p-AlGaIn cladding layer. The dark spots disappeared at 940°C , indicating that reducing the p-AlGaIn growth temperature suppressed thermal degradation of the QWs. This phenomenon occurs in epitaxial green LDs, except that the p-AlGaIn growth temperature must be lowered further to suppress thermal degradation of green QWs [22,43,44].

Internal optical loss

Reducing internal loss is also critical for LDs to reduce threshold currents and increase slope efficiencies [21]. Mg-doped layers are the dominant absorption origin in GaN-based LDs because high Mg doping concentrations in the level of 10^{19} cm^{-3} are usually used in p-type layers due to the low ionization ratio of Mg acceptors. The absorption is primarily from acceptor-bound hole absorption [51,52]. Blue LD structures have been numerically designed and experimentally optimized to reduce internal losses by reducing the overlap between the optical field and p-doped layers [75]. By increasing the thickness of an un-doped upper InGaIn waveguide layer from 20 to 110 nm, the internal loss was reduced from 60

Figure 5 Fluorescence images of blue LD wafers with p-AlGaIn cladding layers grown at temperatures of (a) 960°C , (b) 950°C and (c) 940°C , respectively. Reprinted with permission from Ref. [22]. Copyright 2020, Chinese Laser Press.

Figure 6 (a) Slope efficiencies and internal losses of LD chips dependent on the thickness of un-doped upper InGaN waveguide layer in the LD structures. (b) Typical P - I curves for LD chips with various thickness of un-doped upper InGaN waveguide layer. Reprinted with permission from Ref. [21]. Copyright 2017, IEEE.

to 10 cm⁻¹, as measured by the Hakki-Paoli method. Fig. 6a shows that the slope efficiencies and the internal losses of LD chips depend on the thicknesses of un-doped upper InGaN waveguide layer in the LD structures. Fig. 6b shows typical power-current (P - I) curves for LD chips with various thickness of un-doped upper InGaN waveguide layer. The slope efficiency was 0.36, 0.58, 1.1, and 1.5 W A⁻¹ for LD chips with 20, 70, 90, and 110 nm thick un-doped upper waveguides, respectively. The LD chips had ridge waveguide structures with ridge length of 0.4 mm, and their coating reflectivities of rear and front facets were 95% and 34%, respectively. The slope efficiency was measured under continuous-wave (CW) operation at room temperature. Eight LD chips were measured to obtain slope efficiency data for each LD layer structure as shown in Fig. 6a. The data points were the averaged values, and the small variations were also plotted in Fig. 6a.

Hole transport and distribution

Holes in GaN-based materials have a large effective mass and a low mobility, making it difficult to electrically inject them into n-side QWs [22,53–55]. The QWs of LDs require uniform carrier injection to achieve population inversion to form optical gain. Therefore, it is important to optimize the structure of the QWs to improve the carrier distribution for the MQW of LDs. Our previous research found that under electrical injection, holes were only distributed in the QW close to p-GaN when the quantum barrier (QB) thickness of the blue LED was 13 nm. Reducing the thickness and/or the height of the QB can improve hole injection to n-side QWs [58,59]. We fabricated two blue LDs with different barrier thicknesses. The P - I curves are shown in Fig. 7. The threshold

Figure 7 P - I curves of blue LDs with different barrier thickness. Reprinted with permission from Ref. [22]. Copyright 2020, Chinese Laser Press.

current density of the thin barrier LD was 1 kA cm⁻² and the slope efficiency was 1.5 W A⁻¹. The threshold current density of the thick barrier LD was 1.4 kA cm⁻² and the slope efficiency was only 0.8 W A⁻¹. This was caused by a more uniform hole distribution in the QWs and a higher carrier injection efficiency for the thin barrier blue LD. Thus, the slope efficiency was higher than that of the thick barrier LD.

Determination of internal parameters

To obtain internal parameters such as internal optical loss (α_i) and carrier injection efficiency (η_{inj}) of our blue LDs, three blue LDs were fabricated with different front-facet-coating reflectivities [22]. The P - I curves are shown in Fig. 8a. The threshold current density of the LD with the front-facet-coating reflectivity of 10% was 2.4 kA cm⁻² and the slope efficiency was 1.6 W A⁻¹, while the

Figure 8 (a) Three P - I curves of blue LDs with different front-facet-coating reflectivities, and (b) the inverse of SE as a function of the inverse of α_m for these three blue LDs, and the red line is a linear fitting. Reprinted with permission from Ref. [22]. Copyright 2020, Chinese Laser Press.

threshold current densities of the LDs with reflectivities of 45% and 82% were 1 and 0.6 kA cm^{-2} , respectively. The slope efficiencies were 1.1 and 0.5 W A^{-1} , respectively. LD mirror loss (α_m) is determined by the front- and rear-facet-coating reflectivities and the cavity length [76]. The relationship of α_i , α_m , η_{inj} , and slope efficiency (SE) of an LD can be described by [77,78]:

$$\frac{1}{\text{SE}} = \frac{q\lambda}{hc\eta_{\text{inj}}} \left(1 + \alpha_i \frac{1}{\alpha_m} \right).$$

Fig. 8b plots the inverse of SE as a function of the inverse of α_m for these three blue LDs, and the red line is a linear fitting. According to the formula mentioned above, the η_{inj} can be obtained from the intercept of the fitting curve, and the α_i can be obtained by the slope of the fitting curve, since electronic charge (q), wavelength (λ), Planck constant (h), and light speed (c) are known constants. By doing so, the α_i and the η_{inj} of the blue LDs were determined to be 6.8 cm^{-1} and 90%, respectively. For blue LDs, Becerra *et al.* [79] reported an α_i of 9 cm^{-1}

and an η_{inj} of 73%, and Ryu *et al.* [80] reported an α_i of 9 cm^{-1} and an η_{inj} of 89%. Kawaguchi *et al.* [81] reported an α_i of 7 cm^{-1} and an η_{inj} of 96% for violet LDs with the undoped upper waveguide thickness of 100 nm. By increasing the undoped waveguide thickness to 1100 nm, they reduced the α_i to 2 cm^{-1} but at the expense of a decreased η_{inj} of 85%.

LD characteristics

Blue LDs were also fabricated and packaged with ridge size of 45 μm wide and 1200 μm long [22]. The LD characteristics were measured under pulse operation at room temperature. The pulse width was 0.4 μs and the repetition rate was 10 kHz to minimize self-heating effects. The red data points in Fig. 9a were the optical powers at different currents (up to 1.7 A because of the limitation of pulsed current source). The threshold current density of the laser was 1 kA cm^{-2} , and the slope efficiency was 1.65 W A^{-1} . The red line was used to extrapolate the optical power to 3 A (current density of

Figure 9 (a) P - I curves of a blue LD with a 45 μm ridge width and a 1200 μm cavity length under pulse (red) and CW (black) operation. (b) I - V curve of the blue LD under CW operation. Reprinted with permission from Ref. [22]. Copyright 2020, Chinese Laser Press.

6 kA cm^{-2}), which was expected to reach 4 W. The black data points were output powers under CW operation, where the threshold current was the same, but the slope efficiency significantly decreased to 1 W A^{-1} . At a current density of 6 kA cm^{-2} , the output power was 2.2 W. The decrease of slope efficiency was caused by poor heat dissipation of the package. The current-voltage (*I-V*) curve is shown in Fig. 9b, where the operating voltage was 5.3 V at a current density of 6 kA cm^{-2} . The internal parameters including optical loss and injection efficiency, and the LD characteristics including threshold current density and slope efficiency, indicated our blue LDs were comparable to the state-of-the-art.

PROGRESS OF GREEN LDs

Fabrication of green LDs is more challenging than that of blue LDs, because more In must be incorporated to extend the emission wavelength to the green range. In addition, the InGa_N/Ga_N MQW active region of green LDs suffers more severe thermal degradation [41,43], In segregation [82,83], and lattice mismatch. Besides, a lower growth temperature of the p-AlGa_N cladding layer than that of blue LDs must be used to suppress the thermal degradation, which results in a relatively high series resistance of green LDs. Extensive efforts have been made to improve the luminescence homogeneity and the efficiency of green LD active region [26,27,84]. In order to suppress the thermal degradation, low temperature growth and p-doping of the p-AlGa_N cladding layer were examined [28,48]. Besides, hybrid green LDs with an indium tin oxide (ITO) cladding layer have been fabricated to further suppress the thermal degradation of the green InGa_N MQW active region. Hence, the performance of hybrid green LDs has been greatly improved [29].

Improvement of luminescent homogeneity

The epitaxial growth of the InGa_N/Ga_N MQW active region with high internal quantum efficiency (IQE) and high differential gain is a key point to realize green LDs with low threshold current. Therefore, green InGa_N/Ga_N MQWs with sharp interfaces and homogeneous luminescence are important to lower the threshold current of green LDs.

Step-flow growth of InGa_N QWs can result in smooth morphologies and thus sharp QW interfaces. Oliver *et al.* [85,86] intensively studied the growth modes of InGa_N on Ga_N, and reported step-flow growth for InGa_N QWs with lower In-content than in green InGa_N QWs. However, green InGa_N QWs with high In-content, which

must be grown at temperature lower than 700°C, usually exhibit two-dimensional (2D) island morphology or 3D morphology [84,85–90]. Increasing defects, related to In segregation, at green InGa_N/Ga_N QW interfaces, could be another reason for the rough interface and low IQEs. [84,91–93]. Therefore, to increase the IQE and improve the luminescence homogeneity, the ways to realize step-flow growth of green InGa_N QWs were studied to suppress the formation of interfacial defects for green InGa_N/Ga_N MQWs.

As shown in Fig. 10a, adatoms either diffuse on the epitaxial growth surface and incorporate into atomic steps, or aggregate and nucleate two-dimensionally depending on the substrate terrace width and adatom diffusion length. Step-flow growth is desirable to obtain high-quality QWs with high emission efficiencies and homogeneity. The growth temperature must be lowered to increase the In-content to around 30% in InGa_N QWs for green emission. Reduced adatom diffusion lengths at lower temperatures resulted in 2D growth and morphologies. Substrates with large miscut angles and thus narrower atomic terraces were used to grow InGa_N QWs with high In-content. As shown in Fig. 10b, we established a relationship between the growth mode and substrate miscut angle, i.e., atomic terrace width. For a higher In-content of InGa_N QW, a larger substrate miscut angle was needed to obtain step flow growth. Fig. 10c and d show the atomic force microscopy (AFM) images of 2D island morphology and step-flow morphology, respectively. The root-mean-square (RMS) roughnesses for $1 \times 1 \mu\text{m}^2$ scan areas were 0.24 and 0.16 nm, respectively. Although the roughness was similar, their origins were different. The former resulted more from the 2D morphology, while the latter resulted more from the atomic step edges because it had a greater two-fold step density. Surface height fluctuation for the 2D islands ranged from 0.6 to 1.2 nm, while the step height was 0.26 nm for the step-flow morphology. Thus, In atoms tended to segregate at the edges of 2D islands where strain relaxation was allowed. Improved luminescent homogeneity was therefore expected for step-flow growth. Temperature dependent photoluminescence (PL) results of the green InGa_N/Ga_N MQWs grown on Ga_N substrates with the step-flow morphologies had an IQE one-fold higher than that of MQWs with 2D island morphologies. Moreover, the room-temperature emission line-width of green InGa_N/Ga_N MQWs was greatly reduced from 166 meV for 2D island morphologies to 131 meV for step-flow morphologies at an emission wavelength of 536 nm.

Green InGa_N/Ga_N MQWs are highly strained and

Figure 10 (a) Schematic of formation of different growth modes and morphologies. (b) Relationship between the growth mode of InGa N QWs and the substrate miscut angle depending on In-content; AFM images ($1 \times 1 \mu\text{m}^2$) of green InGa N QW samples grown on *c*-plane GaN substrates with miscut angles of (c) 0.20° , and (d) 0.48° . Reprinted with permission from Ref. [27]. Copyright 2017, AIP Publishing LLC.

must be grown at low temperatures with a large lattice mismatch. This causes a high density of trench defects at the interface, as shown in Fig. 11a. These defects greatly lower their emission efficiency of green InGa N/GaN MQWs [91–93], and are a challenge for green LDs with constant temperature growth (1T growth) of InGa N/GaN MQW active regions, i.e., the same growth temperature for InGa N QWs and GaN QBs, as schematically shown in Fig. 11b. The formation of trench defects is closely related to In segregation, and can be eliminated by the thermal annealing process in which, QWs are grown at 700°C and then the temperature is ramped to 850°C to desorb excess In at the growth surface. The QBs were grown at 850°C to avoid the formation of trench defects, i.e., two temperature growth (2T growth), as schematically shown in Fig. 11b. However, the thermal annealing process resulted in high density of voids ($2.8 \times 10^8 \text{ cm}^{-2}$) at the surface of the QWs, producing a rough QW/QB interface, as shown in Fig. 11c. Besides, thermal annealing process will result in In composition fluctuations caused by In desorption, both of which resulted in broadening of emission spectra. Therefore, an approach to suppress the formation of trench defects rather than thermal annealing process was examined to reduce the emission linewidth of the green InGa N/GaN MQW active region [94]. It was found that

the density of trench defects in green InGa N/GaN MQWs could be suppressed by an order of magnitude by increasing the V/III ratio from 13,000 to 26,000, as shown in Fig. 11a and d, and could be further reduced to $5 \times 10^5 \text{ cm}^{-2}$ by growth on a GaN substrate [94]. Hence, high quality green InGa N/GaN MQWs with 1T growth were demonstrated, as shown in Fig. 11e. A scanning transmission electron microscopy (STEM) image in Fig. 11f of green InGa N/GaN MQWs with 1T growth indicated a very sharp interface. The optical quality of green InGa N/GaN MQWs was thus greatly improved, as indicated by an increased PL intensity and nonradiative recombination lifetime and decreasing spectra full width at half maxima (FWHMs) [94]. Fig. 12 shows a comparison of spectral FWHMs vs. emission energy, reported by various research groups. It revealed that the FWHMs of 113 meV at an emission wavelength of 530 nm for our green InGa N/GaN MQWs was the best reported value so far.

Suppression of thermal degradation

During subsequent p-type AlGa N cladding layer growth, InGa N MQWs tend to degrade in crystalline quality and emission efficiency, and even decompose due to instability at temperatures for p-AlGa N growth [40–44].

Figure 11 (a) AFM image of green InGaN/GaN MQWs with V/III ratio of 13,000. (b) Schematic of 1T growth and 2T growth. (c) AFM image of green InGaN/GaN MQWs from 2T growth. Reprinted with permission from Ref. [84]. Copyright 2017, The Optical Society. (d) AFM image of green InGaN/GaN MQWs with V/III ratio of 26,000. Reprinted with permission from Ref. [94]. Copyright 2019, The Japan Society of Applied Physics. (e) AFM and (f) STEM images of green InGaN/GaN MQWs from 1T growth.

Figure 12 Spectral FWHMs vs. emission energy for green and yellow GaN-based LEDs. The red solid circle is our electroluminescence (EL) FWHMs of 2T grown green LD [84], the red solid star is our PL FWHMs of 1T grown green MQWs [94], the black solid squares are LEDs grown on Si substrates from Nanchang University [95], and other data are PL for LD structures grown on GaN from Nichia [96], EL for LEDs grown on sapphire from Toshiba [97], PL for an LD structure grown on GaN from Osram [42], EL for an LD structure grown on (20 $\bar{2}$ 1) plane GaN substrate from Sumitomo [32], and EL for an LD structure grown on (20 $\bar{2}$ 1) plane GaN substrate from UCSB [98,99].

Thermal degradation is more severe in green LD structures with high In-content in the InGaN/GaN MQW active region. As shown in Fig. 13a, there was a high density of dark regions with different grey scales in micro-PL images of LD wafer, which indicated no or weak

Figure 13 Micro-PL images of a green LD structure with p-AlGaIn grown at (a) 940°C and (b) 900°C. STEM images of (c) non-degraded and (d) degraded regions. Reprinted with permission from Ref. [43]. Copyright 2013, AIP Publishing LLC.

emission caused by the thermal degradation. The different grey scales may indicate that the decomposition occurred in the top or bottom QW simultaneously. To mitigate the thermal budget imposed on InGaN QWs, the growth temperature of p-type layers was lowered by 40°C, and homogeneous luminescent image was obtained, as shown in Fig. 13b. It was noted that the degradation of

InGaN QW was suppressed because of the reduced thermal budget during p-type growth. Fig. 13c shows STEM images of the non-degraded region, which featured continuous and homogeneous InGaN QWs. Bright precipitates and black voids were observed in STEM images of the degraded region, as shown in Fig. 13d. The degradation was caused by thermal decomposition of In-rich InGaN initiated at the In-rich InGaN clusters located at the surface of InGaN QW [43]. Metallic In clusters were generated as a result of the decomposition. By suppressing the thermal degradation, the emission efficiency of the green LD structure was greatly improved [43].

Carbon impurities in the p-AlGaN:Mg cladding layer

To suppress thermal degradation of green InGaN/GaN QWs with high-In-content during the growth of p-type AlGaN cladding layers [6,43], p-type cladding layers were usually grown at a temperature lower than optimal temperature which was higher than 1000°C for AlGaN. However, p-type AlGaN growth at a low temperature often had high resistivity due to increased defects and impurity incorporations [100–103]. Because of the chemical activity of Al, the AlGaN layer grown by MOCVD usually contained higher carbon impurity concentrations

than GaN especially when the growth temperature was reduced below the optimal growth temperatures. The dependence of carbon concentration on growth conditions was examined [28,48]. As shown in Fig. 14a, when the p-type AlGaN cladding layer was grown at a lower temperature to avoid the thermal degradation of the active region, the incorporation of carbon impurities was enhanced. However, by adopting a lower growth rate and a higher growth pressure, carbon concentrations in p-type AlGaN could be suppressed to less than $1 \times 10^{17} \text{ cm}^{-3}$, as shown in Fig. 14b and c. A correlation between carbon concentration and electrical properties of AlGaN:Mg was obtained. When the carbon concentration was higher than a certain level in the order of 10^{17} , there was a clear tendency that the hole concentrations decreased and the resistivity increased with the carbon concentration, as shown in Fig. 14d. Based on the analysis of the charge neutrality equation, carbon impurities acted as the main compensating donors in AlGaN:Mg. The carbon impurity concentration also decreased as the substrate misorientation angle increased, which was attributed to reduced step motion velocity for epitaxial growth on substrates with larger misorientation angles [104].

Spontaneous recombination IQEs were estimated *via* temperature-dependent PL measurements to evaluate the

Figure 14 Dependence of growth conditions on carbon incorporation: (a) growth temperature, (b) growth pressure, and (c) growth rate. (d) Hole concentration and resistivity vs. carbon concentration. Reprinted with permission from Ref. [48]. Copyright 2015, The Japan Society of Applied Physics.

Figure 15 Spontaneous recombination IQE data for our green LD structures and LEDs. Data from Nanchang University were calculated using external quantum efficiency data assuming an extraction efficiency of 70% [95,109]. The red rhombus points are data of our conventional green LDs [26] and hybrid LDs with ITO cladding layer. Other data are from Toshiba [97,110] and UCSB [111,112]. Our data are measured *via* PL for LD structures grown on free-standing GaN substrates, while the literature data were measured *via* EL from LEDs grown on Si or sapphire substrates.

quality of MQW active regions in green LD structure. As shown in Fig. 15, the IQEs of conventional green LD structures with 500 nm p-AlGaIn cladding layers were 59%, 39% and 29% for samples with emission wavelengths of 523, 540 and 554 nm, respectively. Because there have been no reports of spontaneous recombination IQEs for LD structures, state-of-the-art results for LEDs are also shown in Fig. 15. It should be noted that our data were measured by PL for LD structures grown on free-standing GaN substrates, while literature data were measured by EL for LEDs grown on Si or sapphire substrates. It can be seen that the IQEs of our conventional green LD structures with metal electrodes were a little lower than the best results reported by Nanchang University, but

were comparable to other reports. Therefore, by improving the epitaxial growth of green InGaIn QWs and adopting low temperature growth and p-doping of p-AlGaIn cladding layer under optimized conditions to suppress carbon impurities, spontaneous recombination IQE of our green LD structures was greatly enhanced. Fig. 16a and b show threshold current density and slope efficiency *vs.* lasing wavelength for green LDs reported in a review paper by Monavarian *et al.* [105]. The threshold current density and slope efficiency of our green LDs were comparable to the state-of-the-art results. Although the dislocation density of LD structures grown on sapphire substrates was two orders of magnitude higher than those grown on GaN substrates, high IQEs of 41.1% at wavelength of 527 nm and 24.3% at wavelength of 615 nm were also obtained by using quantum dots as gain medium [106,107]. This enabled low-threshold green vertical-cavity surface-emitting lasers (VCSEL) under electrical injection [108].

Hybrid green LDs with ITO cladding layers

ITO is conductive and transparent in the visible spectrum range and has been widely used as an electrode in GaN-based LEDs. Its absorption coefficient is two orders of magnitude lower than that of metals [113–115], and its refractive index is around 2 [116], which is much lower than that of the p-AlGaIn cladding layer. It therefore provides sufficient optical confinement for the laser cavity. ITO can be deposited around 300°C or lower. Thus, replacing a p-AlGaIn cladding layer with ITO can reduce the high-temperature growth time of the p-AlGaIn cladding layer, and thus reduce the thermal budget imposed on green InGaIn QWs. Moreover, using ITO as the cladding layers also allows a lower Al-content p-AlGaIn cladding layer to be used in hybrid LD structures. This enhances the electrical properties of the cladding layer

Figure 16 (a) Threshold current density *vs.* lasing wavelength for green LDs, and (b) slope efficiency *vs.* lasing wavelength for green LDs. Reprinted with permission from Ref. [105]. Copyright 2018, John Wiley & Sons.

Figure 17 (a) *P-I* curves of ITO LDs with 300 nm p-Al_{0.035}Ga_{0.965}N cladding layer and conventional LDs, and (b) *I-V* curves of two different LDs.

including hole concentration and conductivity [28,48]. Therefore, the hybrid GaN-based LDs using ITO as p-cladding layer is promising to improve the performance of GaN-based LDs. There have been a few reports about the design and fabrication of hybrid GaN-based LDs using ITO as a cladding layer. However, the performance is only comparable to LDs with conventional structures [117–120].

The hybrid green LDs with ITO cladding layers were designed and fabricated to suppress thermal degradation of green InGaIn QWs. The performance of hybrid green LDs was greatly improved and the LD structure is shown in Fig. 2. A 200 nm ITO layer was used to replace part of the p-AlGaIn cladding layer. ITO is also a p-electrode, replacing the normal Pd/Pt/Au metal.

The spontaneous recombination IQEs of green LD structures with ITO cladding layer was estimated *via* temperature-dependent PL measurements. The excitation light source was a 405 nm laser and the excitation power density was 2.4 kW cm⁻². The IQE is the ratio of the integrated intensity at room temperature to that at 10 K, assuming that nonradiative recombination is frozen and thus IQE is 100% at 10 K. As shown in Fig. 15, the IQE of hybrid green LD structure with a thinner p-AlGaIn increased to 76% at wavelength of 523 nm. Hence, reducing the p-AlGaIn thickness and its growth time further suppressed the thermal degradation of green QWs [40–44].

Hybrid green LDs with ITO cladding layers were measured by probing LD bars under pulsed operation at room temperature. The *P-I* curves for ITO LDs and conventional LDs are shown in Fig. 17a. The threshold current density of a conventional LD was 5 kA cm⁻², and the slope efficiency was 0.16 W A⁻¹; whereas the threshold current density of ITO LDs with a p-Al_{0.035}Ga_{0.965}N cladding layer was 1.6 kA cm⁻², and the slope efficiency was 0.2 W A⁻¹. Our simulation indicated that the con-

finement factors of these two samples were almost the same. The simulated total internal loss of the sample with 300 nm p-Al_{0.035}Ga_{0.965}N cladding layers was 8.2 cm⁻¹, which was 2.3 cm⁻¹ lower than that of conventional LD sample. This reduction in internal loss was not so significant to account for the reduction of the threshold current density and the increase in the slope efficiency. From our previous report, the improvement of ITO LDs with a p-Al_{0.035}Ga_{0.965}N cladding layer was attributed to the reduced non-radiative recombination centers of QWs and increasing hole concentration and injection efficiency [29]. Fig. 17b shows a comparison of *I-V* curves for ITO LDs and conventional LD under CW electrical injection. With a decrease of p-AlGaIn cladding layer thickness and Al-content, the LD operation voltages decreased. The operation voltage of the ITO LD with the 300 nm p-Al_{0.035}Ga_{0.965}N cladding layer was 0.6 V lower than that of the conventional LD at a current density of 4 kA cm⁻², which was attributed to the reduced series resistance due to reduced thickness and Al-content of p-AlGaIn cladding

Figure 18 *P-I* curve of ITO LDs with a 300 nm p-Al_{0.035}Ga_{0.965}N cladding layer. The front facet coating was 40%.

layer.

Because the threshold current density of the ITO LD with a $\text{p-Al}_{0.035}\text{Ga}_{0.965}\text{N}$ cladding layer was as low as 1.6 kA cm^{-2} , it enabled a reduction in the front facet coating reflectivity to 40%, and an increase in the ridge size to $15 \mu\text{m}$ wide and $1200 \mu\text{m}$ long. The P - I curve of the ITO LDs was measured under pulsed operation as shown in Fig. 18. The threshold current density was 4.4 kA cm^{-2} and the slope efficiency was 0.62 W A^{-1} . The output power was as high as 500 mW at the current density of 9 kA cm^{-2} , which was typical for GaN-based green LDs.

CONCLUSIONS

In summary, the challenges and progress of GaN-based blue and green LDs were reviewed from the aspects of epitaxial growth and layer structure design. Due to large differences in lattice constants and growth conditions among InN, GaN, and AlN, much effort must still be made to improve the quality of InGaN MQW gain medium for blue and especially green LDs. The p-type doping profile, conditions and layer structure are critical for the reduction of internal losses and also to mitigate the degradation of InGaN MQWs. In addition, hole injection is a key issue for GaN-based LDs.

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Aiqin Tian is a postdoctoral researcher in Suzhou Institute of Nano-Tech and Nano-Bionics, Chinese Academy of Sciences (CAS). She earned her doctoral degree from the University of Chinese Academy of Sciences in 2017. Her research focuses on MOCVD growth of III-nitride materials and devices.

Lei Hu is a PhD candidate at the School of Nano-Tech and Nano-Bionics, University of Science and Technology of China. He received his Bachelor degree (2016) from Soochow University. His research focuses on the fabrication and characterizations of GaN-based blue and green laser diodes.

Jianping Liu is a professor at Suzhou Institute of Nano-Tech and Nano-Bionics, CAS. He earned his doctoral degree from the Institute of Semiconductors, CAS in 2004. He worked at the Lab of Optoelectronics Technology at Beijing University of Technology from 2004 to 2006. He did postdoctoral research in the Department of Electrical Engineering at Georgia Institute of Technology from 2006 to 2010. His research interests include MOCVD growth, GaN-based materials and devices.

GaN基蓝光与绿光激光器

田爱琴^{1†}, 胡磊^{1,2†}, 张立群¹, 刘建平^{1,2*}, 杨辉^{1,2}

摘要 GaN基激光二极管(LD)将半导体LD的波长扩展到可见光谱和紫外光谱范围, 因此有望被广泛用于光钟等量子技术、生物医疗仪器、激光显示、照明和材料加工等领域。尽管它们与GaN基发光二极管(LED)基于相同的III氮化物材料, 但是蓝光和绿光LD面临更大的挑战。在本文中, 我们从外延生长和结构设计的角度对GaN基蓝光和绿光LD面临的挑战和进展进行了回顾总结。InN、GaN和AlN之间的晶格常数和生长条件差异很大, 因此需要进行深入研究来提高蓝光, 尤其是绿光LD的InGaIn/GaN多量子阱(MQW)增益介质的材料质量。p型掺杂分布, 生长条件和器件结构对减少内部损耗并抑制InGaIn MQW的热退化至关重要。此外, 空穴注入也是GaN基LD面临的关键问题。