ALGaN/GaN heterostructures with asymmetric graded-index separate confinement heterostructure for electron-beam pumped UV lasers

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Abstract: We present a study of undoped AlGaN/GaN separate confinement heterostructures designed to operate as electron beam pumped ultraviolet lasers. We discuss the effect of spontaneous and piezoelectric polarization on carrier diffusion, comparing the results of cathodoluminescence with electronic simulations of the band structure and Monte Carlo calculations of the electron trajectories. Carrier collection is significantly improved using an asymmetric graded-index separate confinement heterostructure (GRINSCH). The graded layers avoid potential barriers induced by polarization differences in the heterostructure and serve as strain transition buffers which reduce the mosaicity of the active region and the linewidth of spontaneous emission.

1. Introduction

There is a high demand of ultraviolet (UV) lasers for applications in the fields of medicine and biotechnology [1], as well as in 3D printing [2] and non-line-of-sight communication [3]. Presently, this spectral range is covered by gas lasers (ArF, KrF, XeF) or lasers based on frequency conversion (Nd:YAG). III-nitride semiconductor laser diodes are promising candidates to provide an efficient semiconductor-based alternative [4,5]. However, current injection is a major problem for implementing AlGaN-based laser diodes. The p-type doping of AlGaN is particularly challenging [6], and the high doping concentrations required to achieve p-type conductivity degrade the material quality and increase absorption losses. This is one of the main reasons why laser diodes emitting below 370 nm show a dramatic increase in threshold current density [7–11].

An alternative approach is to pump the semiconductor structure with a high energy (5-20 keV) electron beam. Its advantages include higher flexibility in the choice of materials for the active medium due to the absence of doping or electrical contacts, as well as higher radiative recombination efficiency since the electrons and holes generated by impact ionization share the same distribution in the active medium. This technology has allowed the fabrication of ZnSe-based pulsed lasers that emit up to 600 W at 535 nm [12]. There are some studies of electron beam pumped AlGaN for the implementation of UV lamps [13–20]. Wunderer et al. demonstrated UV lasing from an AlGaN/GaN heterostructure containing InGaN quantum wells (QWs) under pulsed electron beam excitation at 77 K [21]. The emission peaked at 384 nm, with a threshold of 174 µA at an acceleration voltage of 18 kV (≈160 kW/cm² assuming the
beam focused to a spot diameter of 50 µm). At the same time, Hayashi et al. showed lasing from an In-free AlGaN/GaN structure under pulsed electron beam excitation at 107 K. A separate confinement heterostructure (SCH) containing a 10-period GaN/AlGaN multi-quantum well (MQW) displayed stimulated emission around 353 nm with a threshold power density around 230 kW/cm² [22]. These demonstrations have opened a new line of research, but new device architectures are required with the prospect of achieving room temperature lasers.

Here, we present a study of undoped AlGaN/GaN SCHs designed to operate under electron beam injection with an acceleration voltage ≤ 10 kV. We discuss the effect of spontaneous and piezoelectric polarization on the carrier diffusion, and demonstrate that the performance is improved using an asymmetric graded-index separate-confinement heterostructure (GRINSCH).

2. Design

Figure 1(a) presents a schematic description of the laser structures under study. The choice of bulk GaN as a substrate aims at minimizing the dislocation density. The active region consists of a 10-period GaN/AlGaN MQW inserted in an AlGaN/AlGaN SCH, where the core (bottom/top inner cladding, BIC/TIC in the figure) has lower Al mole fraction than the outer claddings (bottom/top outer cladding, BOC/TOC in the figure). The waveguide is induced by the variation in the refraction index between the cladding layers. In some samples, the alloy transition between the AlGaN cladding layers is gradually switched in a so-called graded layer, with the ternary alloy composition varying linearly along the growth axis.

The design of the structures was carried out taking into account both optical and electrical considerations: (i) the bottom cladding layer must be thick enough to prevent waveguide losses through the absorbing GaN substrate, (ii) the penetration depth of the electron beam in the structure imposes a limit to the thickness of the upper layers, and (iii) the MQW must emit at the targeted wavelength. Therefore, on the one hand, we studied the confinement of the optical mode in the waveguide using a commercial finite-element analysis software (Comsol Multiphysics) with the values of refractive indices extracted from [23,24]. On the other hand, we conducted Monte Carlo simulations using the CASINO software to predict the path of the electron beam with a targeted operation voltage of 10 kV. Finally, for the design of the MQW geometry, the electronic band profiles were modelled in one-dimension using the Nextnano³ 8-band k·p self-consistent Schrödinger-Poisson solver [25], with the material parameters described in [26]. The structures were considered pseudomorphic on the GaN substrate, and the spontaneous and piezoelectric polarization, as well as the band structure deformation potentials, were taken into account. The thicknesses of QWs and barriers were tuned to attain the targeted operation wavelength. The simulations were performed incorporating an electron density of 10¹⁸ cm⁻³ to mimic operation conditions.

The final architectures, summarized in Table 1, were obtained after several iterations of optical and electronic simulations. As an example, Fig. 1(b) presents the simulation of the transverse-magnetic (TM) polarized optical mode confined in the SCH for sample S1, and Fig. 1(c) shows the predicted trajectory of the electrons in the same sample. The calculations show that the vast majority of carriers are absorbed in the active region for an acceleration voltage of 10 kV. In comparison with the design of Hayashi et al. [22], here the thickness of the upper layers were reduced to operate at a lower acceleration voltage (10 kV instead of 15 kV), and the waveguide core was also smaller, to improve the optical confinement. S1 presents an optical confinement factor of 7%, slightly higher than that reported by Hayashi et al. [22] in spite of our thinner QWs. In the case of samples S2, S3, and S4, the confinement factors are smaller due to the even narrower QWs.
Fig. 1. (a) Schematic view of the laser structure. The following layers are grown sequentially on top of GaN substrate: bottom outer cladding (BOC), bottom inner cladding (BIC), MQWs active region, top inner cladding (TIC) and top outer cladding (TOC). (b) Simulation of the TM optical mode distribution through the SCH for sample S1. (c) Simulation of the electron beam trajectory in the structure S1. The central panel shows the trajectory of the electrons generated by an electron beam under 10 kV. The panel on the right side shows the electron energy loss profile for various acceleration voltages.

Table 1. Description of samples under study (thickness and Al content of the layers, following the general design in Fig. 1(a)) and their optical confinement factor (OCF).

|       | S1          | S2          | S3          | S4          |
|-------|-------------|-------------|-------------|-------------|
| TOC   | 47 nm Al<sub>0.15</sub>GaN<sub>0.85</sub>N | 44.4 nm Al<sub>0.2</sub>GaN<sub>0.8</sub>N | 26.4 nm Al<sub>0.2</sub>GaN<sub>0.8</sub>N | 44.2 nm Al<sub>0.3</sub>GaN<sub>0.7</sub>N |
| Graded| 35.2 nm    | 48.7 nm    |             |             |
| TIC   | 63 nm Al<sub>0.07</sub>GaN<sub>0.93</sub>N | 59.5 nm Al<sub>0.07</sub>GaN<sub>0.93</sub>N | 41.4 nm Al<sub>0.1</sub>GaN<sub>0.9</sub>N |             |
| QWs   | 10×(2.8 nm GaN) | 10×(1.4 nm GaN) | 10×(1.3 nm GaN) | 10×(1.3 nm GaN) |
|       | /11.4 nm Al<sub>0.07</sub>GaN<sub>0.93</sub>N | /9.8 nm Al<sub>0.07</sub>GaN<sub>0.93</sub>N | /9.7 nm Al<sub>0.1</sub>GaN<sub>0.9</sub>N | /9.7 nm Al<sub>0.1</sub>GaN<sub>0.9</sub>N |
| BIC   | 37.6 nm Al<sub>0.07</sub>GaN<sub>0.93</sub>N | 35.5 nm Al<sub>0.1</sub>GaN<sub>0.9</sub>N | 17.6 nm Al<sub>0.1</sub>GaN<sub>0.9</sub>N | 17.7 nm Al<sub>0.1</sub>GaN<sub>0.9</sub>N |
| Graded| 35.2 nm    | 35.4 nm    |             |             |
| BOC   | 376 nm Al<sub>0.15</sub>GaN<sub>0.85</sub>N | 355 nm Al<sub>0.2</sub>GaN<sub>0.8</sub>N | 332 nm Al<sub>0.2</sub>GaN<sub>0.8</sub>N | 334 nm Al<sub>0.2</sub>GaN<sub>0.8</sub>N |
| OCF   | 7%         | 4.1%       | 3.7%        | 3.8%        |

3. Experimental

The laser structures described in Table 1 were synthesized on free-standing, non-intentionally doped, n-type conductive GaN(0001) substrates using plasma-assisted molecular-beam epitaxy, with the growth conditions described elsewhere [27]. The substrate temperature was \( T_S = 720 \) °C and the growth rate was 0.5 monolayers per second (ML/s). Note that in wurtzite GaN and AlGaN, the thickness of 1 ML is approximately 0.25 nm.

The periodicity of the MQW structures was analyzed by high resolution X-ray diffraction (HRXRD) in a Rigaku SmartLab diffractometer. Additional structural studies were conducted on a thin lamella prepared by focused ion beam, using high-angle annular dark-field (HAADF) scanning transmission electron microscopy (STEM) performed on a FEI Thémis microscope operated at 200 kV.
Cathodoluminescence (CL) experiments were carried out in a FEI Inspect F50 field-emission SEM equipped with an iHR550 spectrometer fitted with an Andor Technology Newton DU940 BU2 spectroscopic charge-coupled device (CCD) camera. The beam spot diameter was ~10 nm on the focal point, the scanning area was a square of 4 µm², the accelerating voltage was varied from 5 to 20 kV.

A set of laser bars with varying resonator length (ranging from 0.5 to 3 mm) were prepared by mechanical cleaving along the (10-10) m-plane of GaN. The facets were uncoated. Photoluminescence (PL) measurements were obtained under pulsed excitation using a Nd:YAG laser (λ = 266 nm, pulse duration = 0.5 ns, repetition rate = 8 kHz) at room temperature. Cylindrical lenses were inserted to shape the laser beam into a 100-µm-wide stripe, and to control the beam uniformity (variation of intensity <10% along the longest cavities). All tests were carried out at room temperature. The PL edge emission was collected by a Jobin Yvon HR460 monochromator equipped with a UV-enhanced CCD camera.

4. Results and discussion

Samples S1 and S2 were designed for emission at 360 nm and 355 nm, respectively. The spectral shift is achieved by varying the Al content of the AlGaN layers and the thickness of the active QWs. After growth, the geometry and structural quality of the samples was validated by HRXRD and STEM. Figure 2(a) shows HRXRD θ-2θ scans around the (0002) reflection of GaN for samples S1 and S2, compared with theoretical calculations using the GlobalFit2 software from Rigaku Co. The calculations used the layer thicknesses indicated in Table 1, and the structure was assumed pseudomorphic on the BOC layer, which was allowed to relax partially (relaxation in the range of 50%). The more intense peak of the diffractogram is attributed to the GaN substrate, and the second highest, located at slightly higher angles, to the TOC and BOC layers. The reflection of the MQW presents several satellites, which allow a precise determination of the MQW period. HAADF-STEM images for sample S2 are shown in Fig. 2(b), where clear contrast corresponds to layers with higher Ga concentration. The zoomed image on the right side reveals a QW thickness of 1.4 nm.

![Fig. 2. (a) HRXRD θ-2θ scan around the (0002) reflection of GaN for samples S1 and S2. Labels identify the reflections originating from the GaN substrate, the BOC and TOC layers and the QWs, with several satellites. Reflections from BIC and TIC layers are not resolved due to their proximity to the QW reflection. Experimental results are compared with theoretical calculations of the diffractograms. (b) HAADF-STEM image of the inner section of the heterostructure in sample S2. On the right side, zoomed image showing 3 QWs.](image-url)
The optical characterization of sample S1 is summarized in Fig. 3. Figure 3(a) shows the variation of the CL spectra as a function of the acceleration voltage, $V_A$. The CL spectra are normalized by dividing by the acceleration voltage and injection current, so that their comparison provides a direct view of the evolution of the conversion efficiency. At low acceleration voltage, the emission is dominated by recombination in the QWs (at 360 nm) with a shoulder at 350 nm that is assigned to recombination in the inner cladding. The outer cladding (emitting at 335 nm) is only visible for $V_A \geq 12$ kV. In view of the intensity of the curves and the ratio of the emission from the QWs and from the inner cladding, the efficiency of the device is expected to be maximum in the range of $V_A = 5-7$ kV.

![Fig. 3. (a) CL spectra of sample S1 at different acceleration voltages. (b) Photoluminescence spectra in semi-logarithmic scale of sample S1 (cavity length = 1 mm) at different pumping power. (c) Emitted photoluminescence intensity (full markers) and FWHM (empty markers) in function of the excitation power density. The grey dashed line represents a linear tendency. (d) Dependence of threshold power density with the cavity length.](image)

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Laser bars with cavity lengths between 0.5 and 3 mm were optically pumped using the 266 nm Nd-YAG laser source at room temperature. Figure 3(b) represents the room-temperature PL emission spectra of a 1-mm-long cavity at various pumping power densities. Laser emission appears at 367 nm above the threshold power density of 192 kW/cm². The red shift of our emission in comparison with Hayashi et al. [22] is explained by the fact that all the experiments in this paper are performed at room temperature. Figure 3(c) presents the variation of the PL intensity and its full width at half maximum (FWHM) as a function of the pumping power density, showing an abrupt superlinear increase of the emission at the threshold together with a drop of the FWHM by several orders of magnitude (note that the measured values are limited by the resolution of the setup, around 1 nm). The variation of the power density threshold for different cavity lengths is studied in Fig. 3(d). The threshold decreases when the cavity length increases. We attribute this behavior to mirror losses in the cleaved $m$-facets. Using a simple model, the laser threshold is proportional to the sum of the losses:
\[ P_{th} \propto \left( \alpha_i + \frac{1}{2L} \ln \left( \frac{1}{R^2} \right) \right) \]  

where \( \alpha_i \) are the internal losses, \( L \) is the cavity length and \( R \) is the reflectivity of one of the facets (considered equivalent). Assuming \( \alpha_i = 20 \text{ cm}^{-1} \), the reflectivity of each facet would be \( R \approx 17\% \), which is a reasonable value for uncoated cleaved facets [21].

The same characterization was carried out for sample S2 (Fig. 4), where the QW thickness was reduced to blue shift the emission. Figure 4(a) shows the variation of the CL spectra as a function of \( V_A \). Similar to S1, the highest efficiency is attained for \( V_A \leq 7 \text{ kV} \). At those voltages, the spectra are dominated by the QW emission at 354 nm. At higher acceleration voltages, a secondary peak assigned to \( \text{Al}_{0.1}\text{Ga}_{0.9}\text{N} \) (inner cladding, at 342 nm) appears, and it becomes dominant for \( V_A > 12 \text{ kV} \).

Figure 4(b) presents the edge PL emission of a 1-mm-long cavity from S2. Laser emission at 355 nm arises at a threshold power density of 210 kW/cm\(^2\). In comparison with S1, the reduction of the QWs widths and the higher Al concentration in claddings resulted in an increase of the emission energy. Figure 4(c) illustrates the enhancement of the intensity and the reduction of the FWHM at the threshold. The evolution of the threshold power density as a function of the cavity length is depicted in Fig. 4(d), showing the same trend as S1.

The intensity of the CL emission at 342 nm in Fig. 4(a) reveals a problem of carrier transfer from the inner cladding to the QWs. This problem becomes evident when looking at the band diagram of S2, depicted in Fig. 5(a). The difference in spontaneous and piezoelectric polarization between the waveguide core and the outer cladding results in a strong band bending that favors electron accumulation at the TIC/TOC interface and hole accumulation at the BOC/BIC interface (red arrows in the figure). The perturbation of the bands hinders carrier...
transfer to the MQW. This leads us to consider the implementation of a graded index of refraction separate confinement heterostructure (GRINSCH), which promotes the diffusion of electrons towards the wells while the waveguiding properties of the structure are preserved [28,29]. The capabilities of the GRINSCH design are generally not used in III-nitride laser diodes since they introduce a perturbation of the doping distribution due to the variation of polarization in the graded layers [30,31]. However, these effects are not relevant in an electron-beam pumped device so that the advantages of graded index layers can be fully exploited. Therefore, the design for S3 (see Fig. 6(a)) is a modification of S2 where graded interfaces are inserted between the inner and outer cladding layers, resulting in the band diagram in Fig. 5(b), with smooth energy transitions at the heterointerfaces (arrows in the figure).

Experimentally, the effect of the GRINSCH is directly noticeable in the CL spectra of S3, shown in Fig. 6(b). The spectra present a main peak at 352 nm assigned to the QWs and a secondary peak at 339 nm attributed to the Al$_{0.1}$Ga$_{0.9}$N inner cladding layers. A third peak appearing at 324 nm under high acceleration voltage is assigned to the emission of the Al$_{0.2}$Ga$_{0.8}$N BOC layer when the energy of the injected electrons is high enough to penetrate deeper than the GRINSCH region. The room-temperature PL spectra presented in Fig. 6(c) shows stimulated emission at $\lambda = 358$ nm with a power density threshold of 180 kW/cm$^2$. The strong improvement in CL results in a moderate improvement of the threshold. This is due to the fact that under optical pumping the excited is limited to the topmost $\approx 100$ nm, i.e. the excitation is much more superficial than in the case of electron pumping. Therefore, the improvement is expected to be larger in the case of electron pumping.

This optical characterization data of S3 represent a significant improvement with respect to S2. The QW/cladding CL ratio is significantly higher than that of S2, which demonstrates the improved carrier collection and leads to a lower threshold. We also noticed that the emission peaks from S3 are significantly narrower than those from S2, although both samples have the same alloy compositions, approximately the same QW thickness and they emit at the same wavelengths. The reduction of the FWHM can be attributed to a lower defect density in the structure with GRINSCH due to the smooth change in the lattice parameter between cladding layers. The comparison of the XRD $\omega$-scan from both samples corroborates the improvement in crystalline quality, since the FWHM of the rocking curve of S3 is roughly 58% that of S2. This result is consistent with the structural studies of Sun et al. [28], who showed that such compositionally graded AlGaN layers may serve as a strain transition buffer which blocks threading defects.

In spite of the improvement, further effort is needed to reduce the carrier recombination rate in the top cladding layers. A closer look at Fig. 6(b) reveals that the QW/cladding CL ratio is lower at low acceleration voltages, which implies that some of the carriers recombine in the TIC without reaching the QWs. To address this issue, we consider the implementation of an
asymmetric GRINSCH in sample S4, with the design illustrated in Fig. 6(d). The top graded layer extends now to the edge of the MQW and the Al mole fraction of the top outer cladding goes up to 30%. The purpose of these modifications, outlined with dashed red circled in Figs. 5(b) and 5(c), is to force carriers generated in the top layers to diffuse towards the active region. Following this design, the CL spectra of sample S4 in Fig 6(e) shows the predominance of the MQW peak at 351 nm. A peak at 323 nm, assigned to the Al$_{0.2}$Ga$_{0.8}$N BOC, appears only at high acceleration voltage, when the carriers penetrate deeper than the collection region defined by the GRINSCH. For operation at $V_A \leq 7$ kV, 99% of the emission stems from the MQW. For comparison purposes, in the case of S3 at the same acceleration voltage, the light emitted from the MQW was 75% of the total emission. The PL spectra of S4 in Fig. 6(f) shows stimulated emission at $\lambda = 353$ nm with a room-temperature threshold power density of 180 kW/cm$^2$.

Fig. 6. (a,d) Schematic view of the laser structures (a) S3 and (d) S4. (b,e) CL spectra at different acceleration voltages for (b) S3 and (e) S4. The CL spectra are normalized by dividing by the acceleration voltage and injection current. (c,f) Photoluminescence spectra in semi-logarithmic scale at different pumping power densities for a 1-mm-long cavity from sample (c) S3 and (f) S4.

It should be noted that the optically-pumped threshold power densities presented here could be decreased by eliminating the top cladding layers. However, this would result in a degradation of the performance under electron pumping, due to the different carrier distribution between an optically-pumped and an electron-beam pumped device. The measurement of the optical threshold for the different SCH designs provides comparative information about the carrier transfer from the top layers to the active region and the optical loses in the waveguides. Our experiments demonstrate that the implementation of a GRINSCH results in a relevant improvement of the collection of carriers generated by impact-ionization while maintaining laser emission at 353 nm with a threshold power density lower than that of the equivalent SCH design.

5. Conclusion

In this work, we presented AlGaN/GaN heterostructures designed to operate under electron beam injection for acceleration voltages $\leq 10$ kV, and displaying laser emission at room temperature under optical pumping. The implementation of a GRINSCH improves the collection of carriers towards the active region without any negative impact in the quality of
the waveguide. Furthermore, the smooth transition between ternary alloys in the compositionally graded layers results in an improved structural quality of the active MQW, thus reducing the threshold compared with the SCH. In addition to that, an asymmetric design of the GRINSCH is introduced, to further improve the transfer of carriers towards the active region. The combination of these modifications could lead to new electron-pumped laser devices with high performance in the UV emission range.

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