Deep-UV optical gain in AlGaN-based graded-index separate confinement heterostructure

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Abstract: We propose AlGaN-based GRaded-INdex Separate Confinement Heterostructure as a candidate for electrically pumped deep-UV semiconductor laser. Strong compositional fluctuations were introduced in the active region (75 nm-thick Al\textsubscript{0.72}Ga\textsubscript{0.28}N film grown by RF Plasma-assisted Molecular Beam Epitaxy) to obtain net modal optical gain peaked at 257 nm in excess of 80 cm\textsuperscript{-1}. We measured an optical gain threshold of 14 µJ/cm\textsuperscript{2}. Because of polarization-doping of the compositionally graded AlGaN regions, which automatically leads to the formation of p-i-n junction, these results pave the way for lasing under electrical injection.

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OCIS codes: (140.4780) Optical resonators; (160.3380) Laser materials; (310.6860) Thin films, optical properties; (320.7100) Ultrafast measurements.

References and links


1. Introduction

The demonstration of a compact, portable, solid-state deep-UV laser is strongly required to enable a number of applications in different fields, such as non-line-of-sight free-space optical
communications, biochemical agent detection, disinfection, and medical diagnostics. GaN alloyed with high AlN mole fraction is the most promising platform for sub-250 nm electrically-injected LEDs and lasers [1, 2]. In the last few years, many groups have investigated AlGaN-based light emitting devices operating below 300 nm. The majority of the reported external quantum efficiencies of LEDs are very low so far (less than 3%) [3–7]. Nevertheless, the ability to grow high quality materials has been proven also at wavelengths down to 235 nm, exhibiting internal quantum efficiency up to 70% at 240 nm [8–12], power efficiency of 40% at 240 nm [13], and, more recently, deep UV LEDs with external quantum efficiency above 10% [14] have been reported. Moreover, some prototypes of optically pumped deep-UV lasers as well as the demonstration of stimulated emission have been reported in the literature [15–27]. The employment of multiple quantum well structures is one of the most effective approaches for obtaining lasing. However, homogeneous quantum wells require high carrier density to invert their population before any stimulated emission process sets in. In order to overcome this issue, the authors have already proposed to take advantage of strong band-structure potential fluctuations to lower the threshold gain and increase the net modal gain [17, 28]. An alternative approach consists in the design of a laser structure capable of enhancing both the carrier and the optical confinement factors in the active region. Semiconductor laser designs have evolved extensively over the last few decades targeting lower lasing thresholds with higher gain efficiency. These important performance indicators were demonstrated via several architectural improvements, which include the Separate Confinement Heterostructure (SCH) laser, the quantum well SCH, and the Graded-Indext SCH (GRINSCH) geometry [29–31]. Unlike conventional heterostructures, in a GRINSCH laser the cladding materials are progressively alloyed so that the band gap of the cladding is reduced to match the band gap of the material with the smallest gap. As a result, the GRINSCH design not only efficiently confines carriers but also enhances the optical confinement factor [29–32].

In this paper, we report the development of an AlGaN-based deep-UV laser structure in the form of a GRadIndex Separate Confinement Heterostructure (GRINSCH), which is capable of better carrier and optical confinement. We demonstrate that our approach is able to efficiently inject the photo-excited electrons and holes into the active layer (AlGaN, 72% Al content, grown by RF plasma-assisted Molecular Beam Epitaxy with strong compositional fluctuation). The combination of the material properties and the designed GRINSCH structure leads to a maximum net modal gain in excess of 80 cm$^{-1}$ measured by the Variable Stripe Length technique with an optical gain threshold of 14 µJ/cm$^2$ in a 75 nm single layer active region device. Furthermore, as discussed elsewhere [32], the band diagram of this AlGaN-
based GRINSCH structure has the form of a p-n junction due to polarization n- and p-type doping of the AlGaN graded regions on either side of the active region. Thus, this structure has the potential for the development of an electrically pumped deep UV semiconductor laser.

2. Methods

The laser structures were grown by RF plasma-assisted MBE on the Si face of (0001) 6H-SiC substrates. The MBE system is equipped with an EPI Unibulb rf plasma source for the activation of nitrogen and traditional effusion cells for the evaporation of Al, Ga and In. The substrate temperature was monitored with a thermocouple, placed in the cavity behind the substrate, as well as with a pyrometer. In general, the pyrometer reading was about 100 °C lower than the thermocouple reading. The substrate preparation and film growth were monitored by reflection high-energy electron diffraction (RHEED). The SiC substrates were first cleaned ex situ in organic solvents, followed by dipping them into a heated pirana etch and then in buffered hydrofluoric acid to remove surface contaminants and oxides. In addition, the substrates were cleaned in situ by exposure to Ga flux at 750 °C for complete coverage, followed by fast heating to 850 °C for Ga desorption. This process was found to lead to a sharp √3×√3 R30 reconstruction in the RHEED pattern, which was attributed to the removal of oxygen, carbon, hydrogen and other chemisorbed or physisorbed impurities through the formation of volatile Ga compounds. The structure consists of a 500 nm AlN cladding layer, followed by a 50 nm Al$_x$Ga$_{1-x}$N linearly graded from $x = 1$ to $x = 0.8$. The active region of the device consists of 75 nm of Al$_{0.72}$Ga$_{0.28}$N film, followed by the 50 nm Al$_x$Ga$_{1-x}$N linearly graded from $x = 0.8$ to $x = 1$ and 100 nm AlN cladding layer (see Fig. 1). Indium was used as a surfactant during the growth of the graded AlGaN layers in order to improve their crystal quality. We have previously reported [32] a density of threading dislocations under these growth conditions of about 10$^6$ cm$^{-2}$. Moreover, we measured a RMS roughness of 0.79 nm on a 2µm x 2µm AFM image. The active region was grown under Ga-rich conditions, a growth mode that promotes the development of compositional inhomogeneities leading to deep band structure potential fluctuations [17, 28, 33–35]. It has been already demonstrated that nanoscale fluctuations are mainly responsible for light emission in these materials [17] and of the inhomogeneous broadening of the AlGaN emission [36, 37]. On the other hand, we note that the employment of Indium as a surfactant removes the compositional inhomogeneities as revealed by cathodoluminescence measurements [28]. As a consequence, these nanoscale fluctuations are not present in the graded composition regions, which have no role on the light emission of the structure. As will be discussed later, the optical mode here is TM polarized and its optical-mode confinement in the GRINSCH double hetero-structure, described in Fig. 1, was determined using a commercially available
simulation tool (Comsol Multiphysics). Figure 2 shows the vertical profile of the index of refraction and optical mode. Refractive indices at an emission wavelength of 257 nm of 2.3766, 2.4684 and 2.70439 - 0.3277 \times (0.8 \leq x \leq 1) were used for the AlN, Al_{0.72}Ga_{0.28}N and graded Al_{x}Ga_{1-x}N layers respectively [38]. As illustrated by the calculated transverse mode profile, the optical mode was well confined by the graded AlGaN film, with an optical confinement factor, \Gamma, in the active region of the device of 32.5\%, which is a significant improvement over the reported 1–3\% for most InGaN and AlGaN MQW-based lasers [39, 40].

The optical properties and gain of the designed laser structure have been investigated through the Variable Stripe Length (VSL) technique. It is a very reliable and widely used method for the measurement of the optical gain coefficient in bulk materials. This approach allows measuring unambiguously the net modal gain coefficient and the entire gain spectrum [41–44]. No special sample preparation is needed; however some experimental conditions need to be carefully verified to avoid any artifacts [45]. Data are analyzed within the 1D amplifier model to determine the net modal gain/absorption coefficient. The sample has been excited with 220 nm laser pulses, obtained by pumping a proper fourth-harmonic generation crystal (Spectra Physics GWU-24FL) with a mode-locked ultra-fast high-power Ti:sapphire laser (SpectraPhysics MaiTai, 150 fs pulse width, 82 MHz repetition rate) operating at 880 nm. The pulse separation (12 ns) is much larger than the typical emission lifetime in these systems (few nanoseconds), while the laser pulses have a temporal width shorter than the PL decay time. The combination of these conditions corresponds to a transient single pulse excitation regime. This is not an uncommon choice for probing the potential of novel laser materials. In particular, Amplified Spontaneous Emission (ASE) and gain in AlGaN systems have always been observed in a single pulse excitation regime [15, 16, 19, 23, 24]. Moreover, it has been demonstrated that optical gain can be observed and measured also under these conditions, which represent a more accurate representation of the intrinsic net gain [46]. The laser pulses were focused on the sample surface through a cylindrical lens forming a stripe whose length can be monitored and adjusted through a blade mounted in a motorized computer-controlled stage. The beam profile along the stripe has been measured through the knife-edge technique, resulting in a height of the stripe of 5 \mu m and a maximum stripe length of 250 \mu m, which provides a homogeneous illumination of the sample. Beyond this area the pumping intensity cannot be considered uniform anymore and VSL gain data should not be trusted [45, 47]. The maximum fluence on the sample is 60 \mu J/cm^2. The ASE has been
collected from the cleaved edge of the sample through an UV-transmitting objective, an UV-transmitting movable analyzer, a computer-controlled f/4 monochromator (Cornerstone 260) with UV-efficient gratings, and a lock-in amplifier (Oriel Merlin) coupled to a UV-optimized photomultiplier tube (Oriel Instruments 77348).

3. Discussion

In Fig. 3 we report the measured edge emission spectrum intensity normalized by the excitation fluence as a function of the wavelength. This normalization better emphasizes the super-linear behavior of the emitted intensity. All the spectra would line up if the edge-emission depended linearly on the pumping fluence. On the other hand, we observe that the intensities of the edge-emission spectra evolve non-linearly with the pumping fluence. This behavior corresponds to the super-linear Amplified Spontaneous Emission (ASE) from the sample. We do not observe any significant spectral shift within the accessible fluency range.

We have also investigated the polarization properties of the ASE emission in order to better understand the origin of the observed luminescence and gain. In the polar plot shown in the inset of Fig. 3, we report the peak intensity as a function of the analyzer angle. 0° corresponds to the s polarization of the emission (TM mode); 90° to the p polarization (TE mode). Intensities have been recorded at the highest pump fluence (60 µJ/cm²) and they are reported on a linear scale. The emission from the GRINSCH structure appears strongly TM polarized. It is well known that the ASE polarization in AlGaN materials depends on a number of parameters, such as the Al content, the strain in the active layer, and its thickness [49–51]. In particular, a turnover from the TE to the TM polarization is expected for an Al content of 60-80%, depending on the actual device structure [50–52]. Our result is consistent with what has been already observed in the literature for similar Al composition [48]. It should be stressed also that the active region in our structure is not a quantum well, due to its...
large thickness, so we do not expect any contribution to the polarization shift due to the strain, contrary to what we have previously observed in quantum wells with similar Al composition [47].

We report in Fig. 4(a) the ASE intensity measured at the fixed wavelength of 257 nm (the peak wavelength) as a function of the pumping fluence. The measured intensity has a clear non-linear trend, and threshold fluence can be established at around 14 µJ/cm². This is strong evidence supporting the occurrence of stimulated emission in our structure. Furthermore, all the spectra exhibit a single peak and no secondary peaks are observed at longer or shorter wavelengths, indicating that no other luminescent centers are present either in the active or in the graded region of the sample. In Fig. 4(b) we report the VSL traces collected at different pumping fluences. Data are normalized for a better comparison. Continuous lines represent the best fitting to the experimental data according to the model of a 1D optical amplifier system. We use this model to determine the net modal absorption/gain coefficient of the material at different pumping fluences for the same detection wavelength (257 nm). First, we should note the different character of the VSL traces with increasing pumping fluence. At the lowest fluence (5 and 12 µJ/cm²), the corresponding VSL trace is concave. The results are similar for lower fluences. As a consequence, the fitting procedure results in a negative term, i.e. modal absorption values. On the other hand, the traces turn into a convex function by only varying the excitation fluence. In particular, we determine for both 40 and 60 µJ/cm² a positive net modal gain value.

In Fig. 4(c) we report the entire set of measured net modal gain values as a function of the pumping fluence. First, it should be noted that a transition from negative to positive values occurs at 14 µJ/cm². The optical transparency threshold indicates the onset of stimulated emission in the amplifier, and the measured value is well consistent with the one determined in Fig. 4(a) from the pump power dependence of the edge emission. Given our transient excitation regime, the effective photogenerated carrier density depends only on the pump fluence E, the absorption coefficient \( \alpha \) and the photon energy \( h \nu \) [46]:

\[
N_{\text{eff}}^\text{tr} = \frac{E \alpha \nu}{h \nu_p}
\]

Considering a typical absorption coefficient of \( 10^5 \text{ cm}^{-1} \) at this wavelength [38], we estimate a density of excited carriers at gain threshold of 1.55 x \( 10^{18} \text{ cm}^{-3} \). This value is quite low for a bulk material in the deep-UV wavelength range (usually in the range of low-\( 10^{19} \text{ cm}^{-3} \) [16, 25, 49]). However, previously reported numbers do not consider the effect of the graded structure in increasing the optical confinement factor and the carrier funneling into the active region of the device. Moreover, it has been demonstrated that using a pulse width much shorter than the PL decay time reduces the optical gain thresholds and increases the net modal gain compared with the quasi-steady state excitation conditions [46]. We note also that, contrary to what is expected for a bulk material, the net modal gain in our sample saturates for a pumping power of 20 µJ/cm² (very close to the optical threshold). For any higher fluences, the net modal gain keeps its value at around 80 cm⁻¹. This trend has already been observed in nanocrystal quantum dots of different materials [52, 53], and it is consistent with the presence of compositional fluctuations in the active layer of the laser structure [28]. The net modal gain eventually starts to decrease at the highest fluences most likely due to the onset of thermal effects.

From the measured net modal gain and the calculated confinement factor, we estimate that the material gain for these bulk Al_{0.72}Ga_{0.28}N films is about 250 cm⁻¹. We have reported previously [17, 28] that Al_{0.72}Ga_{0.28}N /AlN multiple quantum wells (MQWs) have a net modal gain of about 125 cm⁻¹. Since the confinement factor for such MQWs is of the order of 1-2%, the estimated materials gain for such MQWs is of the order of 6,000 to about 12,000 cm⁻¹. The superior gain properties of the MQWs compared to bulk films of similar composition are
attributed to better carrier confinement in the MQWs, which prevents them from diffusing and recombining at non-radiative recombination centers. Based on this discussion, we anticipate that the proposed GRINSCH structure will have even higher optical gain if the active region is made of a single or multiple AlGaN quantum wells.

4. Conclusions

In conclusion, we have designed a graded-index laser double heterostructure with AlGaN in the active region to enhance the optical confinement of the structure. The devices were grown by plasma-assisted MBE on the Si face or (0001) 6H-SiC substrates. We have demonstrated large optical gain at room temperature in the deep-UV wavelength range from this structure. Optical gain is demonstrated by super-linear and polarized ASE emission and quantified by VSL analysis that shows clear switching from negative (i.e., absorption) to positive gain values above a characteristic threshold pump fluence of 14 µJ/cm². These results pave the way to the development of sub-250 nm electrically-injected laser structures.

Acknowledgments

The work was partially supported by NSF (ECCS-1408364) and the Defense Advanced Research Projects Agency CMUVT Program (PM: Dr. John Albrecht) under subcontract from Photon Systems Inc. (U.S. Army Cooperative Agreement No. W911NF-11-1-0034).