InGaN laser diodes fabricated by molecular beam epitaxy

MBE 成長による InGaN 系半導体レーザ

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Abstract

Semiconductor nitrides have many applications for optoelectronic devices; particularly, blue-violet laser diodes (LDs) are required for blu-ray optical disc systems. Molecular beam epitaxy (MBE) is a well-established technique for depositing III-V heterostructures, its success exhibited by the MBE-growth of many commercial infrared LDs. However, MBE-growth of nitrides is much more difficult, because providing enough nitrogen atoms at the growth surface, sustaining the high growth temperatures in addition to finding the right growth parameters have proved to be very challenging. We reported the world's first InGaN LDs by MBE in 2004, demonstrating that those problems can be solved and the capability of MBE to produce high-quality optoelectronic devices. Here, we review the significant progress made in the development of nitride LDs by MBE at Sharp Laboratories of Europe. We report on MBE-grown 405 nm InGaN LDs on freestanding GaN substrates and on sapphire templates, with threshold current-densities <10 kA/cm^2 and threshold voltages <10 V, approaching state-of-the-art values. We also demonstrate quasi-cw pulsed operation and report on our efforts to achieve room-temperature cw operation.

窒化物半導体はオプトエレクトロニクス分野で幅広く応用されているが，特に青紫色半導体レーザは光ディスク：BD（Blu-ray Disc）用光源として不可欠である。分子線エピタキシー（MBE）法はIII-V族ヘテロ構造成長方法として既に確立された手法であり，MBE法による赤外レーザの量産も報告されている。しかしながらMBE法による窒化物の成長は，高い成長温度を維持した状態で成長面に十分な窒素原子を供給する必要があり，かつその状態で適切な成長パラメータを探し当てる必要があることから，遥かに難しい挑戦的な研究テーマであった。筆者らは2004年にMBE法としては世界初のInGaN系半導体レーザを発表した。このことは，上記課題が克服可能であることを示すものであり，MBE法が高品質なオプトエレクトロニクスデバイス実現のための有効な手段であることを実証するものである。本稿ではシャープヨーロッパ研究所におけるMBE法による窒化物半導体レーザの研究成果について概説する。筆者らは窒化ガリウム（GaN）基板，およびサファイアテンプレート基板上にMBE法で波長405nmのInGaN系半導体レーザを成長し，閾値電流密度10kA/cm^{2}以下，閾値電圧10V以下を実現し，最先端の水準に近づきつつある。また疑似CW動作にも成功したこと，室温連続発振に向けた取り組みについても述べる。

INTRODUCTION

InGaN blue-violet laser diodes have been the subject of in-tense research in the last ten years, particularly driven by their use in high-density optical storage systems such as Blu-ray. Since the first demonstration of blue-violet laser diodes in 1995 [1], the epitaxial growth of InGaN based laser diodes and light emitting diodes (LEDs) has been dominated by the metal-organic vapor phase epitaxy

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(MOVPE) technique [2, 3]. Molecular beam epitaxy (MBE) with its fine control of growth parameters and capability for in-situ growth monitor-ing is a well-established technique for depositing III-V optoe-lectronic devices, with many commercial infrared laser diodes and LEDs grown this way. However, until recently, MBE had not been successful in producing high-quality InGaN based LEDs and lasers. Despite considerable efforts world-wide, the output power of MBE-grown LEDs had been limited to less than 1 mW [4]. Significant progress in the MBE growth of nitrides has been demonstrated in the last year, with the first InGaN laser diodes grown by MBE [5] and high-power InGaN LEDs grown by MBE with 3.75 mW optical output power at 20 mA current [6] reported by the authors. Subsequently MBE-grown InGaN laser diodes have been reported by an independent group [7]. The first InGaN laser diodes grown by MBE operated under pulsed current conditions at 0.01% duty cycle, with a room-temperature threshold current density of 30 kA/cm^2 and a threshold volta-ge of 33 V [5]. These results were comparable with the first MOVPE-grown InGaN laser diodes and demonstrated the potential of MBE growth for nitride optoelectronic devices. The devices were grown on sapphire templates and subse-quent improvements in growth and fabrication enabled a reduction in threshold current density to 22 kA/cm^2 [8]. These encouraging results opened up the question whether MBE will be able to produce material of high enough quality for room-temperature continuous-wave (cw) laser diodes. Achieving cw operation of MBE-grown laser diodes would be of fundamental interest as well as of commercial signifi-cance. In order to obtain cw-operation at room temperature, further significant reductions in operating current and operat-ing voltage are required, so as to achieve an electrical power dissipation of about 1.5 W or less [9].

In this paper, we report on further substantial improve-ments in material growth and device processing, resulting in threshold current densities of 7.7 kA/cm^2 on sapphire tem-plates. We also report on the MBE-growth of InGaN laser diodes on freestanding GaN substrates, leading to 7 kA/cm^2 threshold current density and high duty-cycle operation of up to 50% at 3.9 °C. The paper is organized as follows. In Sec-tion 1, we describe the laser diode growth by MBE. Section 2 discusses laser structure, fabrication and experimental results for laser diodes grown on sapphire templates; while in Section 3 laser fabrication and experimental results for laser diodes grown on freestanding GaN substrates are presented.

1. LASER DIODE GROWTH BY MOLECULAR BEAM EPITAXY

The laser diodes were grown in an Oxford Instruments V80 MBE system using Ammonia (NH_3) as the source of nitro-gen [5]. The group III elements as well as the n-type dopant were evaporated from gallium, aluminum, indium and silicon solid source effusion cells. Organometallic bis(cyclopentadienyl) magnesium (Cp2Mg) was used as the source of magnesium atoms for p-type doping. Two types of substrates were used for the growth. The first type of sub-strate consists of 10 µm of silicon-doped GaN grown by MOVPE on a sapphire substrate, and is commercially avail-able from Lumilog. These template substrates have a disloca-tion density of 8x10^{7}cm^{-2}. Laser diodes were also grown on a second type of substrate, n-type (0001)-face freestanding GaN substrates, commercially available from Sumitomo Electric Industries Ltd. The threading dislocation density of the freestanding GaN substrates is less than 1x10^{6}cm^{-2} in the areas where the laser waveguides are fabricated. The laser diode structure grown on template and freestanding substrates is the same and is grown according to the following proce-dure. Before growth, molybdenum is deposited on the back-side of the substrates, allowing them to be radiatively heated to the required growth temperature. The laser structure is then grown directly onto the substrate without the use of any low temperature buffer or nucleation layer. The most recent struc-ture of the laser diodes is shown in **Fig. 1** and consists of a separate confinement heterostructure with a multiple quantum well active region. A 260 nm layer of silicon-doped GaN was first grown to bury any residual contamination present on the substrate surface.

This was followed by a 1070 nm silicon-doped $Al_{0.12}Ga_{0.88}N$/GaN short period superlattice cladding layer with 2.6 nm period, and 60 nm of silicon-doped GaN optical waveguide region. Then the laser active region was grown, consisting of 15 nm $Al_{0.11}Ga_{0.89}N$; four undoped, 3 nm thick, In0.1Ga0.9N quantum wells separated by 15 nm thick undoped GaN barriers; and 5 nm $Al_{0.15}Ga_{0.85}N$. The p-doped region consisted of 70 nm of magnesium-doped GaN optical waveguide and a 500 nm magnesium-doped $Al_{0.12}Ga_{0.88}N$/GaN short period superlattice cladding layer with 2.6 nm period; followed by a 20 nm magnesium-doped GaN contact layer. The average aluminum content of the cladding layers was measured to be 6% using X-ray diffrac-tion. Typical growth temperatures were 1000 °C for the GaN and AlGaN layers and 660 °C for the InGaN

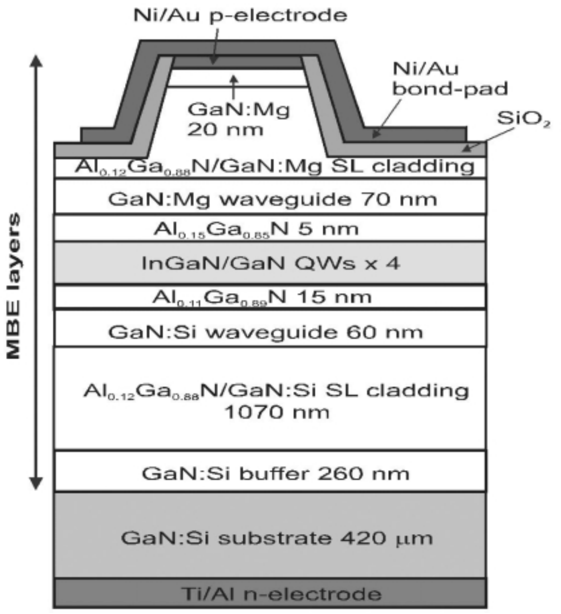

Fig. 1 Structure of the InGaN multiple quantum well (QW) laser diodes as grown on freestanding GaN substrates.

quantum wells. No post-growth thermal annealing to activate the p-type dopant was necessary due to the low H2 content in MBE growth. The nominal quantum well and guiding layer thick-nesses were confirmed by transmission electron microscopy (TEM) [8] and X-ray diffraction.

2. LASER DIODES ON TEMPLATE SUB-STRATES

A. Laser structure and fabrication

After MBE-growth of the laser diode structure on a tem-plate substrate, ridge waveguide lasers were fabricated. Due to the sapphire-based substrate dry-etched facets and a top-side n-contact have to be used. The wafers were processed using the following method. A Ni/Au p-contact electrode was de-posited by thermal evaporation. Electron cyclotron resonance (ECR) plasma etching with a Cl_2/$SiCl_4$/SF_6/Ar chemistry was then used to etch the ridge waveguides, with a width of 2 – 5 µm. An insulating layer of SiO_2 was deposited, serving as an etch mask for 500 to 1500 µm long mesas. The mesas were ECR-etched to the n-doped template substrate to a depth of ~2 µm, thus forming the laser facets. Windows aligned with the waveguides were etched into the SiO_2 in order to contact the p-electrode. A Au bond-pad was subsequently evaporated on top of the SiO_2, contacting the p-electrode. An evaporated Ti/Al contact in between the laser mesas formed the n-type electrode. The contacts were annealed for 5 min at 520 °C in nitrogen to reduce their resistance. **Fig. 2** shows a SEM micrograph of one facet of the fully processed laser diode. The facet roughness was estimated to be <20 nm by SEM. Highly reflective facet coatings were not used for the devices fabricated on template substrates.

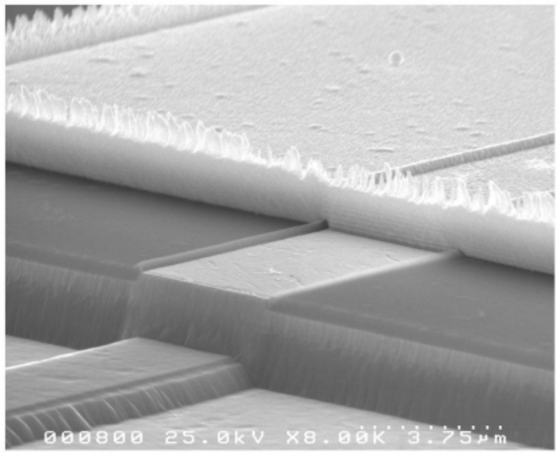

Fig. 2 SEM image of ECR dry-etched laser diode facet.

B. Experimental results for lasers on template substrates

Fig. 3 shows the output power versus injection current (L-I) characteristic typical for our first InGaN laser diodes grown by MBE [5]. It also shows the typical forward bias voltage versus injection current (V-I) characteristics. The waveguide dimensions were 5 µm x 1000 µm, and the clad-ding layers consisted of bulk $Al_{0.08}Ga_{0.92}N$. The devices were electrically operated at room temperature under pulsed current conditions (200 ns pulse width, 0.01% duty cycle). There is a clear threshold in the L-I curve at a lasing threshold current density of 30 kA/cm^2, where the operating voltage was ~33 V. Lasing is also demonstrated by the abrupt narrowing of the emission spectrum above threshold, as indicated in **Fig. 4**. Here stimulated emission with a linewidth of less than 0.2 nm was observed at a wavelength of approximately 400 nm. We subsequently reduced the threshold current density by 25% from our first reported lasers by improving the quantum efficiency in the active region through optimization of the growth temperatures [8]. We were able to reduce the threshold current density even further through the following improve-ments. First, we replaced the bulk $Al_{0.08}Ga_{0.92}N$ cladding layers with superlattices. The n-cladding layer consisted of a 1.1 _m Si:($Al_{0.12}GaN_{0.88}$/GaN) short period

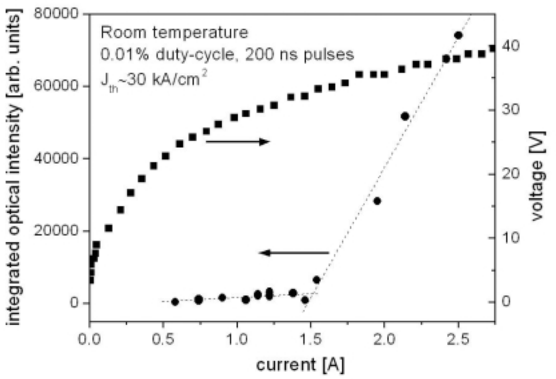

Fig. 3 L-I and V-I characteristics of first MBE-grown laser diodes.

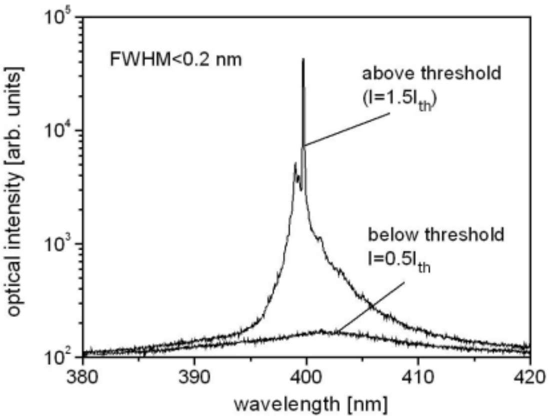

Fig. 4 Emission spectra of typical laser diode below and above threshold.

superlattice (SL) with a 2.6 nm period; the p-cladding layer consisted of a 500 nm Mg-doped ($Al_{0.12}GaN_{0.88}$/GaN) short period superlattice with 2.6 nm period. Secondly, we used longer devices (5 µm x 1600 µm) to reduce the effect of mirror loss. Finally, fur-ther optimization of the growth process led to a further im-provement in quantum efficiency in the active region. **Fig. 5** shows the L-I curve of a typical laser diode with these improvements. The devices were measured at room tempera-ture under the same pulsed conditions as before (200 ns pulse width, 0.01% duty cycle). The threshold current density was reduced to 7.7 kA/cm^2, a considerable improvement and comparable to MOVPE-grown laser diodes on templates without facet coating [10]. The operating voltage at threshold was 25 V, with the reduction being in part due to the reduced threshold current.

3. LASER DIODES ON FREESTANDING GAN SUBSTRATES

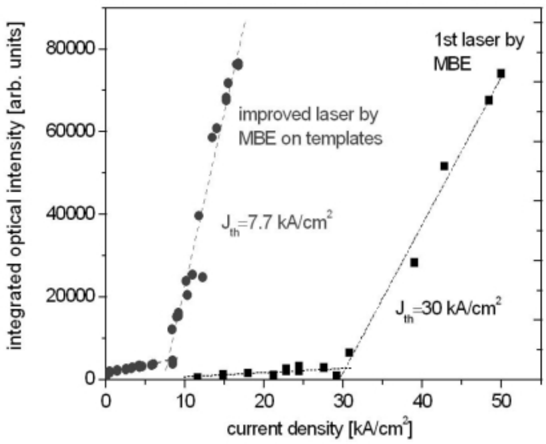

Fig. 5 L-I characteristics of improved laser diode grown by MBE on template substrate, compared to first laser diode on template substrate.

A. Laser structure and fabrication

In this section we report on the fabrication of MBE-grown laser diodes on the freestanding GaN substrates described in Section 1. These substrates have several advantages over sapphire substrates, including high-quality cleaved facets, higher thermal conductivity and backside n-contacts [11]. As outlined in Section 1, we grow the same laser diode structure on the freestanding GaN substrates as on the sapphire tem-plates. The wafers were then processed into ridge waveguide structures using the following method. The Ni/Au p-contact electrode was deposited by thermal evaporation. Electron cyclotron resonance (ECR) plasma etching with a $Cl_2/SiCl_4/SF_6$/Ar chemistry was then used to etch the 2 - 3 µm wide ridge waveguides. An insulating layer of SiO_2 was deposited, into which windows aligned with the waveguides were etched in order to contact the p-electrode. A Au bond-pad was subsequently evaporated on top of the SiO_2, con-tacting the p-electrode. An evaporated Ti/Al contact on the backside surface of the substrate formed the n-type electrode. The wafer was then cleaved into 900 - 1100 µm long laser bars. AFM measurements show that the cleaved facets are usually atomically flat over at least tens of square microme-ters. On some of the laser bars, highly reflective coatings consisting of three or four pairs of quarter-wave Si_3N_4/SiO_2 were applied to each facet after cleaving. The coatings were deposited in a plasma-enhanced chemical vapor deposition system at 300 °C and their reflectivity measured to be ~50% (three pairs) and ~90% (four pairs) at a wavelength of 400 nm. The laser bars were mounted on copper heat sinks to improve thermal management. Several wafers were grown and fabricated into laser devices.

B. Experimental results for lasers on freestanding GaN substrates

Typical L-I and V-I characteristics for laser diodes on freestanding GaN substrates are shown in **Fig. 6**. The laser diodes have superlattice cladding layers as described above and the waveguide dimensions were 3 μm x 1100 μm. A three pair facet coating was applied to the front facet, and a four pair coating to the rear facet. The devices were tested at room temperature under pulsed current conditions (200 ns pulse width, 0.01% duty cycle). The L-I curve gives a thresh-old current density of 7 kA/cm^2 at an operating voltage of ~10V. Lasing is confirmed by the narrow spectrum above threshold. These results compare well with the above results for MBE-grown laser diodes on template substrates, demon-strating the successful transfer of the laser diode growth proc-ess from the sapphire templates to the freestanding GaN substrate.

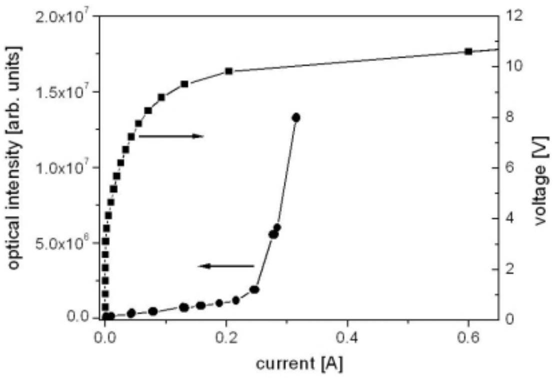

Fig. 6 L-I and V-I characteristics of laser diodes grown by MBE on freestanding GaN substrate.

In order to achieve the targeted electrical power dissipation of 1.5 W or less, both the operating current and the operating voltage must be minimized. Subsequently, we have im-proved the threshold voltage of our devices by optimizing the p-doping level in the structure. Compared to a voltage of up to 11 V at 8 kA/cm^2 measured previously, a 25% reduction to 8 V has been achieved with the improved Mg-doping. This brings the voltage closer to state-of-the-art threshold voltages measured on MOVPE laser diodes [12, 13].

C. Operation of MBE-grown laser diodes at high duty cycles

While the results reported so far were obtained under low duty cycle conditions, we now report on laser measurements under high duty cycle operation. For the experiment we used a freestanding GaN laser diode bar with 2 μm x 915 μm waveguide dimensions and improved p-doping as above. The laser diodes were mounted on a temperature-controlled sink and the measurements performed at 3.9 °C. Facet coatings were not employed for this measurement. The electrical pump pulse width was kept constant at 500 μs, increasing the threshold current density to 11.7 kA/cm^2 for duty cycles of less than 10% (see **Fig. 7**). The threshold current was 215 mA, and the power dissipation at threshold 1.8 W. The increase in threshold compared to the threshold measured with 200 ns pulses can be attributed to an increase in the active region temperature during the 500 μs pulse [14].

While keeping the pulse width constant, the duty cycle was then increased to analyze the effect of long-term heating on the device. **Fig. 7** shows the resulting threshold current versus duty cycle curve. One can see from the graph that the threshold current density keeps increasing as the duty cycle is increased beyond 10%. We ascribe the threshold increase to increased overall heating of the device, as the generated heat cannot be removed from the active region in between the pulses [14]. Lasing stopped at a maximum duty cycle of 50% due to device failure. We believe this is the highest reported duty cycle operation for InGaN laser diodes grown by MBE. The duty cycle measurements demonstrate the progress that has been made since the demonstration of the first MBE-grown InGaN laser diode. With the use of highly reflective facet coatings we expect to reduce the threshold current and electrical power dissipation further, permitting operation at even higher duty cycles.

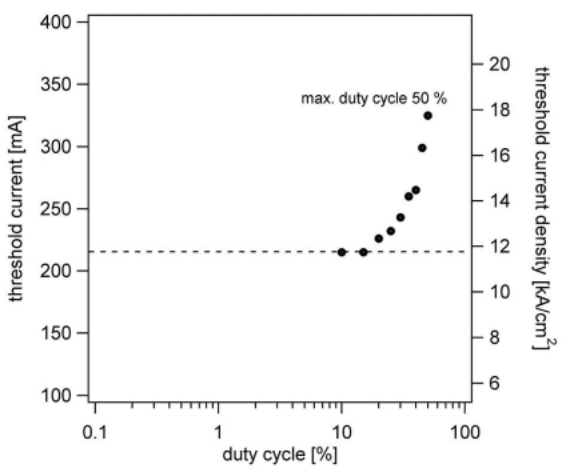

Fig. 7 Threshold current as a function of duty cycle. Measurement on freestanding GaN laser without facet coating at 3.9 °C. The electrical pulse width was kept constant (500 μs).

Conclusions

In summary, MBE has been successfully used to produce InGaN multiple quantum well laser diodes. Laser diodes were grown on sapphire templates and freestanding GaN substrates, and pulsed threshold current densities of 7.7 and 7.0 kA/cm^2 were demonstrated, respectively. The results demonstrate the capability of MBE for the growth of high-quality InGaN laser diodes. Operation at 50% duty cycle at 3.9 °C was shown for laser diodes on freestanding GaN, reflecting the progress made in reducing threshold current density as well as operat-ing voltage. Further optimization of the p-type doping in the AlGaN cladding region and further optimization of growth temperatures are now being investigated with an aim to re-duce the operating voltage and threshold current density in order to obtain continuous-wave operation.

Note added in proof: After submission of this paper, the authors have demonstrated room temperature continuous-wave operation of InGaN laser diodes fabricated by molecular beam epitaxy. Technical details are published in [15].

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