Stimulated emission at 288 nm from silicon-doped AlGaN-based multiple-quantum-well laser

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Abstract: We demonstrated stimulated emission at 288 nm from a silicon-doped AlGaN-based multiple-quantum-well (MQW) ultraviolet (UV) laser grown on sapphire. The optical pumping threshold energy density of the UV laser was 64 mJ/cm², while lasing behavior was not observed in undoped AlGaN MQWs. This means silicon doping could effectively reduce the lasing threshold of UV lasers, and the mechanism was studied showing that the silicon-doped AlGaN MQWs had a 41% higher internal quantum efficiency (IQE) compared with the undoped one. The transmission electron microscopy characterization showed that silicon doping explicitly improved the crystallographic quality of MQWs. Calculation of the polarization charge in the MQWs further revealed that the advantage of better structure quality outweighed the reduction of internal polarization field by Si doping for the IQE enhancement and successful stimulated emission.

OCIS codes: (140.3610) Lasers, ultraviolet; (140.5960) Semiconductor lasers; (230.5590) Quantum-well, -wire and -dot devices.

References and links
1. M. Kneissl, T. Kolbe, C. Chua, V. Kaehler, N. Lobo, J. Stellmach, A. Knauer, H. Rodriguez, S. Einfeldt, Z. Yang, N. M. Johnson, and M. Wever, “Advances in group III-nitride-based deep UV light-emitting diode technology,” Semicond. Sci. Technol. 26(1), 014036 (2011).
2. Y. Muramoto, M. Kimura, and S. Nouda, “Development and future of ultraviolet light-emitting diodes: UV-LED will replace the UV lamp,” Semicond. Sci. Technol. 29(8), 084004 (2014).
3. Y. Taniyasu, M. Kasu, and T. Makimoto, “An aluminium nitride light-emitting diode with a wavelength of 210 nanometres,” Nature 441(7091), 325–328 (2006).
4. S. F. Chichibu, A. Uedono, T. Onuma, B. A. Haskell, A. Chakraborty, T. Koyama, P. T. Fini, S. Keller, S. P. Denbaars, J. S. Speck, U. K. Mishra, S. Nakamura, S. Yamaguchi, S. Kamiyama, H. Amano, I. Akasaki, J. Han, and T. Sota, “Origin of defect-insensitive emission probability in In-containing (Al,In,Ga)N alloy semiconductors,” Nat. Mater. 5(10), 810–816 (2006).
5. J. Simon, V. Protasenko, C. Lian, H. Xing, and D. Jena, “Polarization-induced hole doping in wide-band-gap uniaxial semiconductor heterostructures,” Science 327(5961), 60–64 (2010).
6. M. Shatalov, W. Sun, R. Jain, A. Lunev, X. Hu, A. Dobrinsky, Y. Bilenko, J. Yang, G. A. Garrett, L. E. Rodak, M. Wabraek, M. Shur, and R. Gaska, “High power AlGaN ultraviolet light emitters,” Semicond. Sci. Technol. 29(8), 084007 (2014).
7. M. A. Khan, M. Shatalov, H. P. Maruska, H. M. Wang, and E. Kuokstis, “III–Nitride UV devices,” Jpn. J. Appl. Phys. 44(10), 7191–7206 (2005).
8. M. Shatalov, J. Yang, W. Sun, R. Kennedy, R. Gaska, K. Liu, M. Shur, and G. Tamulaitis, “Efficiency of light emission in high aluminum content AlGaN quantum wells,” J. Appl. Phys. 105(7), 073103 (2009).
9. Z. Lochner, T.-T. Kao, Y.-S. Liu, X.-H. Li, M. Mahbub Satter, S.-C. Shen, P. Douglas Yoder, J.-H. Ryou, R. D. Dupuis, Y. Wei, H. Xie, A. Fischer, and F. A. Ponce, “Deep-ultraviolet lasing at 243 nm from photo-pumped AlGaN/AlN heterostructure on AlN substrate,” Appl. Phys. Lett. 102(10), 101110 (2013).
1. Introduction

Ultraviolet (UV) lasers are of great interest owing to their potential applications in high-density optical storage, biochemical detection, disinfection, medical curing, and micromachining [1]. Compared with traditional laser sources such as excimer lasers and quadrupled Nd:YAG lasers, semiconductor lasers have advantages of low power consumption, compact size and eco-friendly nature, etc [2]. Among different semiconductors, AlGaN alloys have a direct and wide band gap range that covers wavelengths from 210 nm [3] to 365 nm [4], making them ideal candidates for the next-generation UV coherent light sources. However, the development of AlGaN-based UV lasers is a challenge, due to the low hole concentration caused by the difficult $p$-type doping [5], as well as the low internal quantum efficiency (IQE) [6] caused by the high threading dislocation density (TDD) [7] and the deleterious effect of polarization charge in MQWs [8].

Recently, several groups reported on optical-pumped room temperature (RT, 295K) sub-300 nm UV lasers [9–13], mainly focusing on TDD suppression methods, including homoepitaxy on native AlN substrates [9, 13], epitaxy lateral overgrowth (ELOG) [10], and GaN inserted layer technology [11] on sapphire substrates. On the other hand, the influence of doping profile on AlGaN-based UV laser was less explored, especially in the MQWs region. Si doping in MQWs has been proved by Nakamura et al. to be essential for InGaN-based laser diodes, in terms of threshold reduction and lifetime extension [14]. However, it remains unclear whether Si doping has the similar advantageous effect on AlGaN-based UV laser.
diodes. In this letter, we applied silicon doping into AlGaN-based MQWs laser grown on the sapphire substrate and demonstrated stimulated emission from the MQWs at a wavelength of 288 nm by optical pumping. No lasing behavior was observed in the counterpart without silicon doping. The impact of the silicon doping in AlGaN-based MQWs was analyzed by cross-sectional transmission electron microscopy (TEM) and temperature-dependent photoluminescence (PL), showing better interface quality and higher IQE, respectively, than the undoped AlGaN-based MQWs.

2. Experiment

Figure 1 shows the schematic diagram for AlGaN-based MQWs UV lasers. Both silicon-doped (Si-doped) and undoped Al0.35Ga0.65N/Al0.45Ga0.55N MQWs were grown on (0001) sapphire substrate, respectively, by using a metal-organic chemical vapor deposition (MOCVD) system with a common growth pressure of 50 Torr. Hydrogen was the carrier gas, while trimethylaluminum (TMAI), trimethylgallium (TMGa), silane and ammonia were precursors. The epitaxial process was started with a 50-nm low-temperature AlN buffer layer at 550 °C, and then the temperature increased to 1200 °C for 1-μm AlN template deposition. AlN/AlGaN super-lattices (SLs) were inserted between the AlN template and the n-Al0.55Ga0.45N layer to release the stress as well as block dislocation propagation [15]. The 3-μm thick n-Al0.55Ga0.45N layer was used as the cladding layer for a high optical confinement factor. The MQWs structure contained 5 pairs of 3-nm Al0.35Ga0.65N wells and 12-nm Al0.45Ga0.55N barriers. For the Si-doped MQWs, the silicon doping concentration was 3 × 10^{17} cm^{-3} in both quantum barriers and wells.

After the growth process, a SiN layer with thickness of 100 nm was deposited by PECVD as a cap layer. Fabry-Perot cavities were fabricated by cleaving the sample along the [11 2 0] a-direction using a 355-nm laser scribe to form m-plane facet, and the cavity length was 1 mm. Facet coating was not applied to the facet. A KrF excimer laser (λ = 248 nm) was used as the optical pumping source. The beam size was 300 μm × 2000 μm. The incidence optical power from the KrF excimer laser can be adjusted by a filter and monitored by a power meter. The pulse width was 20 ns and the repetition frequency was 5 Hz for sufficient heat dissipation at RT.

3. Results and discussion

Before the MQWs growth, the crystalline quality of the AlN template was evaluated by X-ray diffraction (XRD) and atomic force microscope (AFM). The full width at half maximum (FWHM) values for (0002) and (10 1 2) reflection were 98.5 arcsec and 617 arcsec, respectively. The densities of screw and edge dislocation were estimated to be 2.0 × 10^{7} cm^{-2} and 4.3 × 10^{9} cm^{-2}, respectively [16]. AFM measurement shows the root-mean-square (RMS)
surface roughness of a 5 μm × 5 μm area is 0.227 nm. Two types of laser structures, the Si-doped MQWs and undoped MQWs, were grown under the same conditions to guarantee the same epitaxial layers quality beneath the AlGaN-based MQWs. To analyze the strain in the epilayers grown on AlN template, reciprocal space mapping of the (105) asymmetric Bragg peak by X-ray diffraction was carried out. As shown in Fig. 2, the n-AlGaN film reciprocal lattice points does not fall exactly on the vertical line that is perpendicular to the Qx axis and include the reciprocal lattice points of AlN buffer. This indicates that n-AlGaN is not fully strained with respect to the underlying AlN materials. Six oblique lines have been introduced into the figure from left to right to show 100%, 80%, 60%, 40%, 20% relaxation, and full strain. The relaxation degree of the epilayers is estimated to be about 60%.

The edge emission spectra were acquired by an Ocean Optics Maya 2000 Pro spectrometer with a resolution of 0.12 nm and a detection range from 200 nm to 420 nm. As shown in Fig. 3, when the optical pumping energy density is 20 mJ/cm², below the threshold power density (Pth), the edge emission spectrum exhibits a broad spontaneous emission at 290 nm with an FWHM of 10 nm (blue dashed line). This spectrum is magnified by five times for better comparison with the lasing spectrum (red line). Beyond the Pth of 64 mJ/cm², a sharp peak at 288 nm turns up with the FWHM of 1.6 nm along with some other longitudinal modes peak at 298 nm and 308 nm. The longer emissions are construed to be other competing lasing modes. The prominent decrease of FWHM along with the increasing excitation power density clearly demonstrated the lasing behaviors in the UV laser bar. Unfortunately, when the pumping energy exceeds the Pth, the intensity of lasing emission decreased. This is attributed to the poor ability of heat dissipation, so the laser was burned even fast if the energy went much higher. Compare with the Si-doped MQWs, however, no lasing behavior was observed in the undoped AlGaN-based MQWs until the laser bars were burned under even higher pumping energy. These results imply that silicon doping could effectively reduce the lasing threshold of AlGaN-based UV lasers. This phenomenon has never been reported for AlGaN-based deep UV lasers. However, similar effect has been reported in GaN-based visible LDs. Nakamura et al had remarkably improved the performance of InGaN-based laser diode by Si doping in the quantum well as well as barrier [14]. This is consistent with our result.
Fig. 3. Stimulated spectrum at wavelength of 288nm at room temperature (295K).

We have evaluated the optical mode profile for our MQW laser structure, and the refractive indices for the AlGaN materials were taken from [17]. Due to the highly asymmetric structure, the waveguide layer could not form mode cutoff efficiently. The optical confinement factor ($\Gamma$) of the fundamental TE mode of the electromagnetic field is 0.38%. The optical field maximum intensity is located far away from the center of the MQW active region. The very low optical confinement factor added much difficulty to achieve lasing of our non-optimized structure. This could explain the no lasing behavior of the un-doped MQWs and the high threshold of the Si-doped ones.

We further carried out structural and luminescence characterization to clarify the physical mechanism of our results. Figures 4(a) and 4(b) show the cross-sectional TEM images of Si-doped and undoped AlGaN MQWs, respectively. According to full thickness of MQWs, the types of MQWs have the same growth rate, which means silicon doping does not have an effect on the growth rate of AlGaN alloy. Si-doped MQWs have clearer and more interrupted interfaces between the quantum barriers and wells than the undoped MQWs due to the enhancement of Al migration by Si doping. The sharp interfaces and the compositional uniformity of individual layers in Si-doped MQWs indicate that there is no obvious Al clustering or compositional fluctuation. Previously, Li et al reported AlGaN/AlN MQWs with Si-doped well showed much sharper fringe peaks characterized by high-resolution X-ray diffraction [18], which provided an indirect evidence of interfacial quality improvement by silicon doping. Here, our TEM results furnish a visible proof of the silicon-assisted interfacial improvement. This is attributed to the surfactant effect of silicon doping [19] in MOCVD epitaxy. The atomic migration on the wafer surface was enhanced by silicon doping to generate better interface in MQWs. Better structural quality of MQWs could help increase the radiative recombination in the quantum wells and reduce the lasing threshold.
Temperature dependent PL measurement was carried out to evaluate the IQE of Si-doped AlGaN MQWs and undoped AlGaN MQWs. A fourth harmonic of YAG: Nd laser ($\lambda = 266$ nm, pulse width = 7 ns, frequency = 5 kHz) with a power density of 15 kW/cm$^2$ was used as the excitation source, and an Princeton Instrument spectrometer (Acton SP2750 with 0.02 nm resolution) was used to record the PL spectra. We assumed that the IQE was equal to the integrated PL intensity at RT (295K) normalized by the maximum value at low temperature (8K). Figures 5(a) and 5(b) shows the integrated PL intensity and peak shift of these two samples as the temperature varied from 8K to 295K. The IQE is 42% for Si-doped MQWs and 29.7% for undoped MQWs, showing 41% improvement. Both of their peak wavelengths moved towards to lower-energy side (red shift) as the temperature increased due to the negative temperature coefficient of bang gap, as modeled by Varshni [20]. The IQE enhancement is firstly attributed to the interface improvement of MQW, as mentioned above.

Si doping introduced polarization screening effect is another possible factor for the IQE enhancement. Therefore, we did calculation of the polarization charge in our MQW structures, based on the parameters provided by [21]. The spontaneous and piezoelectric polarization charges are estimated to be 0.0048 C/m$^2$ and 0.0030 C/m$^2$, respectively. The total
polarization charge density at every III-N hetero-interface is $4.9416 \times 10^{12}$ cm$^{-2}$, and the total polarization charge leads to an internal field of 0.9977 MV/cm$^2$. The electron provided by the Si dopant is $4.5 \times 10^{11}$ cm$^{-2}$ at most, assuming that Si dopant is fully ionized. This value is much smaller compared to the intrinsic polarization charge density. Consequently, we consider the field screening to be a minor effect. The advantage of better structure quality outweighs the reduction of internal polarization field by Si doping.

4. Summary

In conclusion, we have studied the influence of Si-doping on AlGaN-based MQW laser grown on sapphire substrate, and demonstrated optically pumped stimulated emission at 288nm, while no lasing behavior was observed for MQWs without Si-doping. To explore the physical mechanism, experimental characterizations and theoretical calculation were carried out. TEM images clearly show that silicon doping could improve the interface quality of MQW. PL results indicate that Si-doped AlGaN MQWs had a 41% higher internal quantum efficiency (IQE) compared with undoped AlGaN MQWs. This great enhancement in IQE is more attributed to the crystallographic factor than the polarization factor brought by Si doping. In one word, Si doping is consider beneficial to achieve efficient deep UV laser diodes. We have already noticed the poor optical confinement in our structure, which resulted in the high threshold. Therefore, further structural optimization and better cavity fabrication process (for example, high high-reflectivity cavity mirror [22]) will be adopted for developing future low-threshold deep UV laser diodes.

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