Deep-ultraviolet lasing at 243 nm from photo-pumped AlGaN/AlN heterostructure on AlN substrate

Zachary Lochner, Tsung-Ting Kao, Yuh-Shiuan Liu, Xiao-Hang Li, Md. Mahbub Satter, Shyh-Chiang Shen, P. Douglas Yoder, Jae-Hyun Ryou, Russell D. Dupuis' , Yong Wei, Hongen Xie, Alec Fischer, and Fernando A. Ponce

Citation: Appl. Phys. Lett. **102**, 101110 (2013); doi: 10.1063/1.4795719
View online: http://dx.doi.org/10.1063/1.4795719
View Table of Contents: http://aip.scitation.org/toc/apl/102/10
Published by the American Institute of Physics

Articles you may be interested in

Low-threshold stimulated emission at 249 nm and 256 nm from AlGaN-based multiple-quantum-well lasers grown on sapphire substrates
Applied Physics Letters **105**, 141106 (2014); 10.1063/1.4897527

Demonstration of an ultraviolet 336 nm AlGaN multiple-quantum-well laser diode
Applied Physics Letters **93**, 241106 (2008); 10.1063/1.3050539

Demonstration of transverse-magnetic deep-ultraviolet stimulated emission from AlGaN multiple-quantum-well lasers grown on a sapphire substrate
Applied Physics Letters **106**, 041115 (2015); 10.1063/1.4906590

Sub-250 nm low-threshold deep-ultraviolet AlGaN-based heterostructure laser employing HfO2/SiO2 dielectric mirrors
103, 211103211103 (2013); 10.1063/1.4829477

Strain dependence on polarization properties of AlGaN and AlGaN-based ultraviolet lasers grown on AlN substrates
Applied Physics Letters **106**, 232101 (2015); 10.1063/1.4922385

Lasing and longitudinal cavity modes in photo-pumped deep ultraviolet AlGaN heterostructures
Applied Physics Letters **102**, 171102 (2013); 10.1063/1.4803689

Deep-ultraviolet lasing at 243 nm from photo-pumped AlGaN/AlN heterostructure on AlN substrate

Zachary Lochner,[1] Tsung-Ting Kao,[1] Yuh-Shiuan Liu,[1] Xiao-Hang Li,[1] Md. Mahbub Satter,[1] Shyh-Chiang Shen,[1] P. Douglas Yoder,[1] Jae-Hyun Ryou,[1,2] Russell D. Dupuis,[1,a)] Yong Wei,[3] Hongen Xie,[3] Alec Fischer,[3] and Fernando A. Ponce[3]
[1]*Center for Compound Semiconductors and School of Electrical and Computer Engineering, Georgia Institute of Technology, 777 Atlantic Dr. NW, Atlanta, Georgia 30332-0250, USA*
[2]*Department of Mechanical Engineering and Texas Center for Superconductivity at the University of Houston (TcSUH), University of Houston, Houston, Texas 77204-4006, USA*
[3]*Department of Physics, Arizona State University, Tempe, Arizona 85287-1504, USA*

(Received 2 January 2013; accepted 5 March 2013; published online 14 March 2013)

Deep-ultraviolet lasing was achieved at 243.5 nm from an $Al_xGa_{1-x}N$-based multi-quantum-well structure using a pulsed excimer laser for optical pumping. The threshold pump power density at room-temperature was 427 kW/cm^2 with transverse electric (TE)-polarization-dominant emission. The structure was epitaxially grown by metalorganic chemical vapor deposition on an Al-polar free-standing AlN (0001) substrate. Stimulated emission is achieved by design of the active region, optimizing the growth, and the reduction in defect density afforded by homoepitaxial growth of AlN buffer layers on AlN substrates, demonstrating the feasibility of deep-ultraviolet diode lasers on free-standing AlN. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4795719]

The germicidal characteristics of deep-ultraviolet (DUV) radiation at $\lambda < 280$ nm have inspired the development of optoelectronic devices such as light emitting diodes (LEDs) and laser diodes (LDs) for applications such as water purification, bio-agent detection, and pathogen sterilization.[1,2] Conventional sources of UV radiation include high pressure mercury lamps and excimer lasers, which suffer the disadvantages of containing toxic materials and low portability. A compact and efficient semiconductor device is, therefore, a desirable alternative. The AlGaInN material system can access the entire UV spectral range from near-(320–400 nm)[3,4] to middle-(280–320 nm)[5,6] and deep-UV (200–280 nm).[7,8] However, the current performance of AlGaInN light-emitting devices is limited and worsens as the wavelength is reduced and the aluminum (Al) mole fraction is increased owing to a reduction of electron and hole concentrations, low injection efficiencies, and a high density of crystalline defects. In particular, threading dislocations act as carrier traps that inhibit radiative recombination thus increasing the power threshold for stimulated emission or even preventing it altogether.[9] Thus, improvements in material quality are required for $Al_xGa_{1-x}N$ ($x > 0.50$) to achieve high-power operation and lasing in the DUV range.

The majority of previously reported devices have been grown heteroepitaxially on non-native substrates such as sapphire or SiC. However, electrical-injection LDs are thus far limited to an emission wavelength of 336 nm due to limitations in effectively doping AlGaN with high Al content.[10] Optically pumped AlGaN multi-quantum well (MQW) structures have been reported for wavelengths as low as 241.5 nm on SiC; however, the threshold power density remains high.[11] While foreign substrates are readily available at reasonable cost, the differences in lattice constant and thermal expansion coefficient with III-nitride materials lead to the formation of high densities of threading dislocations and even cracking for high Al-content $Al_xGa_{1-x}N$.[12] Several strain-relief techniques including superlattices[13] and epitaxial lateral overgrowth schemes[14] have been reported to improve performance through reductions in defect density, yet stimulated emission below 300 nm remains difficult to achieve and is rarely reported.

Native AlN substrates have recently become available to produce DUV optoelectronic devices. Prepared from high-quality bulk crystal by sublimation-recondensation of AlN powder, these substrates enable homoepitaxial growth of AlN buffer layers,[15] leading to a reduced threading dislocation density in the subsequent $Al_xGa_{1-x}N$ active region. The elimination of thermal mismatch between the substrate and epitaxial layers also enhances the growth and performance of the device by eliminating cracks that form during the thermal cycle and cool-down. There have been a handful of reports of growth on native-AlN substrates for both electrically and optically pumped UV devices,[16,17] but efficiencies remain low and thus continued development is underway for better performance at lower wavelengths.

In this study, we have used an Al-polar (0001) native AlN substrate grown by physical vapor transport. Prior to growth, the AlN substrates were prepared in a 3:1 H_2SO_4:H_3PO_4 solution at 90 °C to remove the native surface oxide.[15] An *in situ* high-temperature ammonia treatment was then used to further etch the oxide to enable efficient AlN growth. The epitaxial structure was grown in an Aixtron 6×2 in. metalorganic chemical vapor deposition (MOCVD) reactor with close-coupled showerhead. Due to the low adatom mobility of Al atoms, a higher temperature and lower V/III ratio are required for AlN compared to GaN to promote two-dimensional growth and smooth surface formation.[18–20] Thus, we have used a relatively high temperature of 1155 °C and low V/III ratio for the

[a)]Author to whom correspondence should be addressed. Electronic mail: dupuis@gatech.edu. Also at School of Materials Science and Engineering, Georgia Institute of Technology, Atlanta, Georgia 30332-0245, USA.

 © 2013 American Institute of Physics

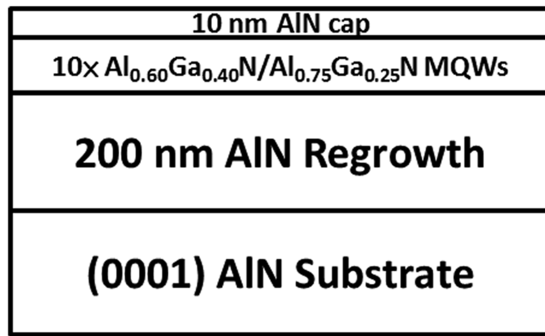

FIG. 1. Cross-section schematic of the DUV AlGaN MQW laser structure.

AlN buffer (<100) and the ternary active region (∼400) for this structure. An even higher growth temperature would be ideal; however, inherent limitations in the reactor design limit us from increasing the temperature further without risking damage to the system.

The epitaxial structure designed for this study was specifically optimized for optical pumping experiments. A cross-section of the layer structure is shown in Figure 1. The active region consists of a 10-period MQW structure, with 3 nm $Al_{0.60}Ga_{0.40}N$ wells and 5 nm $Al_{0.75}Ga_{0.25}N$ barriers, between a 200 nm AlN regrowth buffer layer and a 10 nm AlN cap layer. The cap layer serves as a surface passivation layer for photo-generated carrier confinement, and also as a cladding layer. Although increasing the thickness of this layer would increase the transverse optical confinement factor of the active region, the absorption losses associated with using a 193 nm ArF excitation laser mandate that this layer remains thin. An excitation source with higher wavelength would allow us to grow thicker cladding layers, transparent to the excitation laser, for improved optical confinement and lower threshold power density. However, the active region would also be transparent to a 248 nm KrF laser, thus, constraining the design parameter space for a sub-248 nm photo-pumped emitter. The omission of any underlying AlGaN layers for the purpose of strain reduction serves to enhance the confinement of the optical field within the active region. Even without such strain-engineered layers, high crystal-quality was still maintained as measured by atomic force microscopy illustrated in Figure 2: terraced step-flow is observed, characteristic of two-dimensional layer-by-layer III-nitride epitaxial growth. The RMS roughness is 0.37 nm and 0.32 nm at $10 \times 10\,\mu m^2$ and $5 \times 5\,\mu m^2$ measurements, respectively, indicating slight roughening during growth on the AlN substrate with RMS roughness of 0.1 nm at $5 \times 5\,\mu m^2$, while maintaining a low defect density.

Following growth, the wafer was thinned to 80 μm by lapping the AlN substrate and a Fabry-Perot cavity was formed using cleaved *m*-plane facets with a 1.23-mm long resonant cavity. Neither high-reflection nor anti-reflection coating was used. The laser bars were optically pumped by an ArF Excimer laser ($\lambda = 193$ nm) with a pulse width of 20 ns at a repetition rate of 10 Hz. The laser beam passes through an optical aperture with width of 0.1 cm and length of 1.27 cm and was illuminated over the surface of the cleaved bars. Attenuators were inserted to vary the pump power and a Glan-Laser α-BBO (α-BaB_2O_4) polarizer was used to measure the polarization of light emission. An optical fiber was placed in the proximity of the *m*-plane facets for spontaneous and stimulated light emission detection (as shown in Figure 3). The photon emission was collected and analyzed using an Ocean Optics Maya 2000 Pro spectrometer with a spectral resolution of 0.2 nm.

The photon emission spectrum at room-temperature with different pumping power densities is shown in Figure 4. The peak emission wavelength was $\lambda = 243.5$ nm with the spectral linewidth reducing to 2.1 nm at the maximum measured pump-power of 620 kW/cm^2. The optical output power as a function of excitation density (*L-L* curve) is shown in Figure 5 demonstrating a distinct threshold characteristic at a threshold pump power density (P_{th}) of 427 kW/cm^2. The stimulated emission output increases linearly with the pumping power density beyond the threshold. The measured P_{th} is about three times lower than previously reported photo-pumped UV lasers grown on 4H-SiC at a similar emission wavelength of 241.5 nm.[11] Measurements were also taken from the c^+ plane (Al-face *c*-plane) surface. The linewidth of the spontaneous emission was 12 nm at 600 kW/cm^2.

The transverse electric (TE) and transverse magnetic (TM) emission spectra from the cleaved laser bar operating above threshold are shown in Figure 6. We observed that the stimulated emission is strongly TE polarized with the degree of polarization (P), defined as $P = (I_{TE} - I_{TM})/(I_{TE} + I_{TM})$, greater than 0.9. It is also noted that the TE mode emission wavelength is about 1.6 nm longer than that for TM mode emission. For AlGaN-based lasers, the polarization of the light emission is predicated to switch from the TE mode to

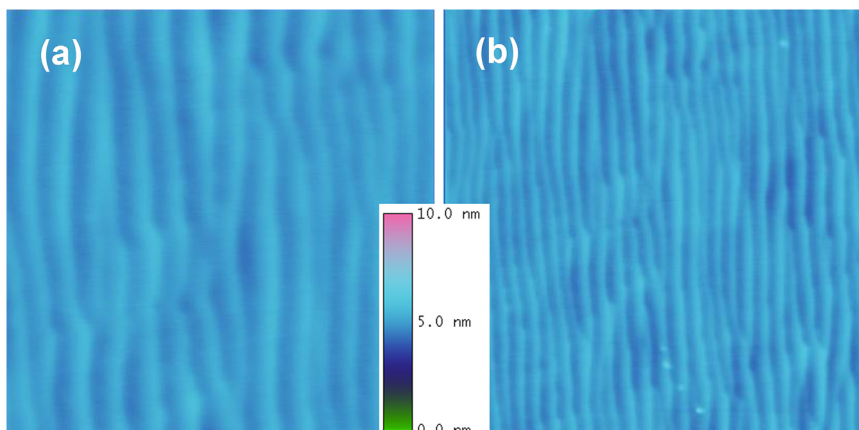

FIG. 2. Atomic-force microscopy measurements of the structure grown on (0001) AlN substrate taken at (a) $5 \times 5\,\mu m^2$ and (b) $10 \times 10\,\mu m^2$.

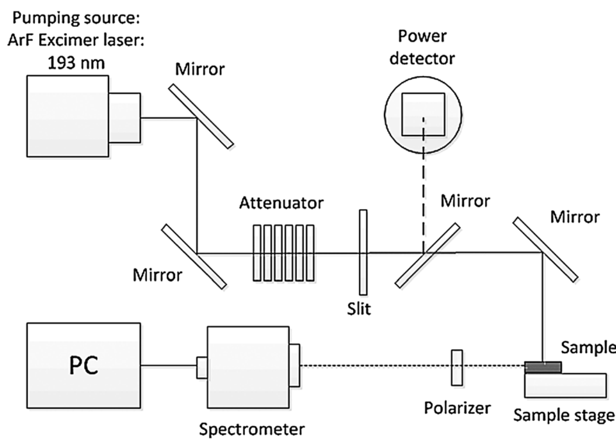

FIG. 3. Schematic diagram of optical measurement system.

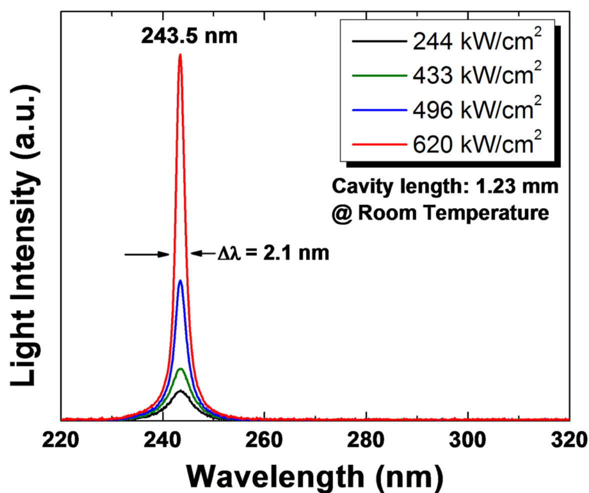

FIG. 4. The laser emission spectra with pump power densities below and above threshold at room-temperature.

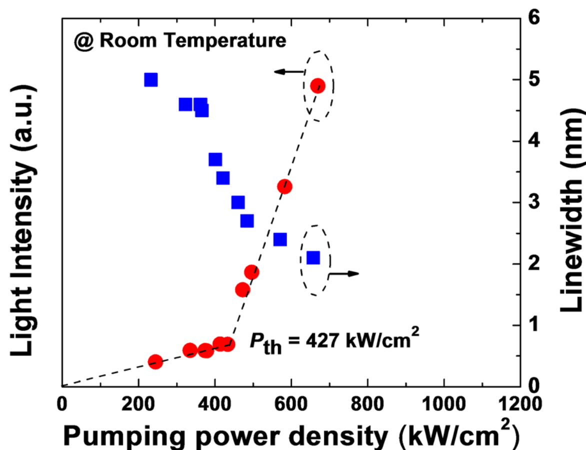

FIG. 5. Light-output intensity and linewidth as a function of the optical pump power density at room-temperature with lasing at $\lambda = 243.5$ nm.

the TM mode as the emission wavelength decreases from near UV to deep UV with corresponding increased AlN mole fraction since the topmost valence band transitions from heavy hole (TE polarization) to crystal-field-split hole (TM polarization).[21,22] Our results (along with those of Refs. 7, 17, and 23) suggest a mixing of these valence bands with a

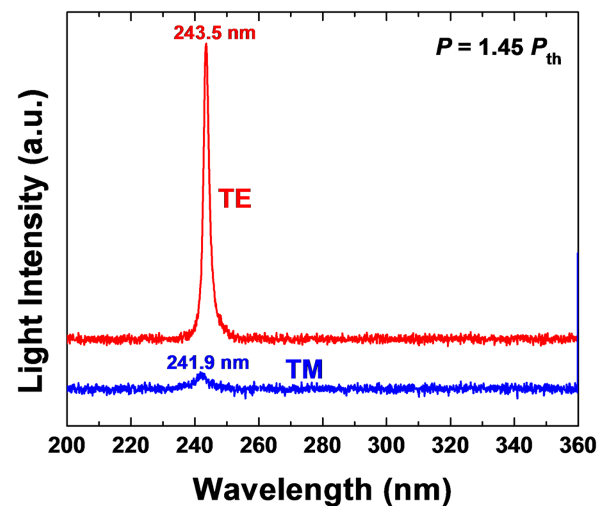

FIG. 6. TE and TM-modes emission spectra for the same laser bar operating above threshold at room-temperature.

dependence on composition and strain state enables TE-mode gain to continue to dominate into the DUV region, while TM-mode sees a modest boost.

In summary, we have used native AlN substrates to grow AlGaN/AlN based MQW heterostructures by MOCVD for DUV stimulated emission. The elimination of thermal mismatch reduction in the strain state between the substrate and the AlN buffer layer yielded reduced defect formation compared to conventional substrates such as sapphire, thus enabling laser action. The peak wavelength demonstrated was at 243.5 nm with a threshold power density of 427 kW/cm^2 and dominating TE polarization mode. These results confirm the viability of AlN substrates for future development of DUV AlGaInN based laser diodes.

This work was supported by the Defence Advanced Research Projects Agency under Contract No. FA2386-10-1-4152. R.D.D. acknowledges additional support of the Steve W. Chaddick Endowed Chair in Electro-Optics and the Georgia Research Alliance.

[1]T. Cutler and J. Zimmerman, Anim. Health Res. Rev. **12**, 15 (2011).
[2]H.-H. Chun, J.-Y. Kim, and K. B. Song, Food Sci. Biotechnol. **19**, 547 (2010).
[3]P. Parbrook and T. Wang, IEEE J. Sel. Top. Quantum Electron. **17**, 1402 (2011).
[4]Y. Ohba, K. Kaneko, H. Katsuno, and M. Kushibe, Appl. Phys. Express **1**, 101101 (2008).
[5]V. Adivarahan, A. Heidari, B. Zhang, Q. Fareed, S. Hwang, M. Islam, and A. Khan, Appl. Phys. Express **2**, 102101 (2009).
[6]S. Fujikawa and H. Hirayama, Appl. Phys. Express **4**, 061002 (2011).
[7]T. Wunderer, C. Chua, Z. Yang, J. Northrup, N. Johnson, G. Garrett, H. Shen, and M. Wraback, Appl. Phys. Express **4**, 092101 (2011).
[8]A. Yasan, R. McClintock, K. Myes, D. Shiell, L. Gautero, S. Darvish, P. Kung, and M. Razeghi, Appl. Phys. Lett. **83**, 4701 (2003).
[9]J. Mickevičius, J. Jurkevičius, K. Kazlauskas, A. Žukauskas, G. Tamulaitis, M. S. Shur, M. Shatalov, J. Yang, and R. Gaska, Appl. Phys. Lett. **100**, 081902 (2012).
[10]H. Yoshida, Y. Yamashita, M. Kuwabara, and H. Kan, Appl. Phys. Lett. **93**, 241106 (2008).
[11]T. Takano, Y. Narita, A. Horiuchi, and H. Kawanishi, Appl. Phys. Lett. **84**, 3567 (2004).
[12]K. Ban, J. Yamamoto, K. Takeda, K. Ide, M. Iwaya, T. Takeuchi, S. Kamiyama, I. Akasaki, and H. Amano, Appl. Phys. Express **4**, 052101 (2011).
[13]J. P. Zhang, H. M. Wang, M. E. Gaevski, C. Q. Chen, Q. Fareed, J. W. Yang, G. Simin, and M. Asif Khan, Appl. Phys. Lett. **80**, 3542 (2002).

[14]H. Yoshida, Y. Yamashita, M. Kuwabara, and H. Kan, Nat. Photonics **2**, 551 (2008).
[15]A. Rice, R. Collazo, J. Tweedie, R. Dalmau, S. Mita, J. Xie, and Z. Sitar, J. Appl. Phys. **108**, 043510 (2010).
[16]M. Kneissl, Z. Yang, M. Teepe, C. Knollenberg, O. Schmidt, P. Kiesel, and N. Johnson, J. Appl. Phys. **101**, 123103 (2007).
[17]T. Wunderer, C. L. Chua, J. E. Northrup, Z. Yang, N. M. Johnson, M. Kneissl, G. A. Garrett, H. Shen, M. Wraback, B. Moody, H. S. Craft, R. Schlesser, R. F. Dalmau, and Z. Sitar, Phys. Status Solidi C **9**, 822 (2012).
[18]H. J. Kim, S. Choi, D. Yoo, J.-H. Ryou, R. D. Dupuis, R. F. Dalmau, P. Lu, and Z. Sitar, Appl. Phys. Lett. **93**, 022103 (2008).
[19]J. R. Grandusky, M. Jamil, V. Jindal, N. Tripathi, and F. Shahedipour-Sandvik, J. Vac. Sci. Technol. A **25**, 441 (2007).
[20]T. Ohba and R. Sato, J. Cryst. Growth **221**, 258 (2000).
[21]H. Kawanishi, M. Senuma, and T. Nukui, Appl. Phys. Lett. **89**, 041126 (2006).
[22]K. B. Nam, J. Li, M. L. Nakarmi, J. Y. Lin, and H. X. Jiang, Appl. Phys. Lett. **84**, 5264 (2004).
[23]W. W. Chow and M. Kneissl, J. Appl. Phys. **98**, 114502 (2005).