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

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Deep-ultraviolet lasing was achieved at 243.5 nm from an AlxGa1−xN-based multi-quantum-well structure using a pulsed excimer laser for optical pumping. The threshold pump power density at room-temperature was 427 kW/cm² 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 λ < 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 AlGaN/N 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 AlGaN/N 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 AlxGa1−xN (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 AlxGa1−xN.12 Several strain-relief techniques including superlattices13 and epitaxial lateral overgrowth schemes14 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 AlxGa1−xN 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 H₂SO₄:H₃PO₄ 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

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AIN 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 excimer 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 x 10 $\mu$m$^2$ and 5 x 5 $\mu$m$^2$ measurements, respectively, indicating slight roughening during growth on the AlN substrate with RMS roughness of 0.1 nm at 5 x 5 $\mu$m$^2$, while maintaining a low defect density.

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

Following growth, the wafer was thinned to 80 $\mu$m 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 $\chi$-BBO ($\chi$-BaB$_2$O$_4$) 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 density ($P_{\text{th}}$) of 427 kW/cm$^2$. The stimulated emission output increases linearly with the pumping power density beyond the threshold. The measured $P_{\text{th}}$ 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.\textsuperscript{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 x 5 $\mu$m$^2$ and (b) 10 x 10 $\mu$m$^2$.\textsuperscript{10}
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). Our results (along with those of Refs. 7, 17, and 23) suggest a mixing of these valence bands with a 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 AIN 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/cm² and dominating TE polarization mode. These results confirm the viability of AlN substrates for future development of DUV AlGaInN based laser diodes.

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