

AlGaN nanocrystals: building blocks for efficient ultraviolet optoelectronics

XIANHE LIU,^{1,2} KISHWAR MASHOOQ,¹ DAVID A. LALEYAN,¹ ERIC T. REID,¹  AND ZETIAN MI^{1,*} 

¹Department of Electrical Engineering and Computer Science, University of Michigan, Ann Arbor, Michigan 48109, USA

²Department of Electrical and Computer Engineering, McGill University, Montreal, Quebec H3A 0E9, Canada

*Corresponding author: ztmi@umich.edu

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AlGaN nanocrystals have emerged as the building blocks of future optoelectronic devices operating in the ultraviolet (UV) spectral range. In this article, we describe the design and performance characteristics of AlGaN nanocrystal UV light-emitting diodes (LEDs) and surface-emitting UV laser diodes. The selective-area epitaxy and structural, optical, and electrical properties of AlGaN nanocrystals are presented. The recent experimental demonstrations of AlGaN nanocrystal LEDs and laser diodes are also discussed. © 2019 Chinese Laser Press

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1. INTRODUCTION

Ultraviolet (UV) light-emitting diodes (LEDs) and laser diodes are important for a broad range of applications, including water purification, disinfection, medical diagnostics, material processing, free-space optical communication, and atomic sensors [1–6]. High wall-plug efficiency, relatively high output power, and stable emission wavelengths are essential for these applications. In addition, UV laser diodes with very narrow spectral linewidths are critical for applications in high-performance atomic clocks and sensors [7,8]. In the past decade, significant progress has been made in UV optoelectronics with the use of AlGaN quantum wells [9–19]. External quantum efficiency (EQE) in the range of 10%–20% has been demonstrated in AlGaN quantum well LEDs operating at ~280 nm [12,16]. However, in a large part of the UV-C spectrum, the EQE of AlGaN quantum well LEDs remains extremely low (well below 1%) [17,20,21]. In addition, there have been no demonstrations of UV vertical cavity surface-emitting laser (VCSEL) diodes with the use of conventional AlGaN quantum wells [22]. The shortest wavelength for electrically pumped AlGaN quantum well edge-emitting lasers is limited to ~336 nm [23]. Some of the critical challenges for achieving high-performance AlGaN UV optoelectronic devices include the presence of large densities of defects and dislocations in the device active region due to the relatively large lattice mismatch (~2.5%) between AlN and GaN. Conventional planar AlGaN structures also exhibit extremely poor p-type conduction due to the very large activation energy of p-type (Mg) dopant (up to ~600 meV) and the formation of extensive compensating defects during epitaxy [24–26]. In addition, light polarization becomes predominantly transverse-magnetic (TM) polarized for Al-rich AlGaN [27], which prevents the

realization of efficient light extraction for LEDs. The strong strain-induced polarization fields and the resulting quantum-confined Stark effect further decrease the device efficiency and the maximum achievable gain for laser diodes.

Recent studies suggest that AlGaN nanostructures hold immense promise to break the efficiency bottleneck of UV optoelectronics [28–38]. Due to the efficient surface strain relaxation, dislocation-free AlGaN nanostructures can be grown directly on foreign substrates including silicon, sapphire, and metals [29,39–48]. It has also been discovered that the formation energy for Al(Ga)-substitutional Mg dopant is significantly reduced in AlGaN nanostructures compared to their planar counterparts [42,49]. Room temperature hole concentration up to $6 \times 10^{17} \text{ cm}^{-3}$ has been measured in AlN nanostructures, which is nearly 7 orders of magnitude larger than that reported in AlN epilayers [25,49]. With the use of such AlGaN nanostructures, UV LEDs and laser diodes operating in the wavelength range of 210–340 nm have been demonstrated [28,31,35,42,50]. The challenge in forming metal contact on nanowires can be addressed by pendeo-epitaxy [36,51]. To date, such AlGaN nanostructures are generally formed spontaneously during epitaxy, which leads to large variations of size, spacing, and surface morphology. To realize high-performance UV optoelectronic devices, it is imperative to achieve AlGaN nanostructures and nanostructure arrays with precisely controlled properties. Such issues can be addressed with the use of the technique of selective-area epitaxy [33,36,52–58]. In this process, the formation of AlGaN nanostructures takes place on a nano-patterned substrate. As such, their size, spacing, and surface morphology can be predetermined and precisely controlled by the underlying nanoscale patterns. The alloy composition and therefore the emission wavelength can also be tuned through variations in the growth

conditions. Through the formation of core-shell heterostructures, detrimental effects such as surface recombination can be minimized or eliminated [33,59,60]. The resulting near-perfect AlGaIn nanocrystals can therefore form the building blocks for next-generation UV optoelectronic devices.

Significantly, through the use of selective-area epitaxy, AlGaIn nanocrystal arrays can form photonic crystal, or meta-material structures with distinct properties and functionality. For example, by coupling in-plane emission to vertical emission, light-extraction efficiency (LEE) of AlGaIn photonic crystal structures can, in principle, reach over 90% from the device top surface for TM-polarized photons, compared to <10% LEE for conventional planar quantum well devices [61]. In addition, surface-emitting UV lasers can be achieved by exploiting the two-dimensional band-edge resonant effect of an AlGaIn photonic crystal [62–65]. At the band edge, the group velocity of light becomes zero, and a stable two-dimensional (2D) cavity mode (standing wave) is formed in the photonic crystal structure. The photonic crystal itself serves as a diffraction grating, and the output beam is emitted from the surface. Previously, an output power >1.5 W and a slope efficiency ~0.66 W/A have been achieved in an InGaAs photonic crystal surface-emitting laser operating at 941.5 nm under continuous-wave operation at room temperature [65]. However, there have been few studies on GaN photonic crystal surface-emitting lasers [66–68]. To date, the only reported electrically injected InGaIn photonic crystal surface-emitting lasers have operated in the blue-violet spectral range ($\lambda \sim 406$ nm) with an extremely large threshold current density (~ 67 kA/cm²) due to the large densities of defects in etched planar structures. Recently, with the use of nearly dislocation-free AlGaIn nanocrystals, electrically injected photonic crystal lasers operating at ~ 369.6 nm have been demonstrated [69].

In this paper, we discuss the design, epitaxy, characterization, and emerging device applications of AlGaIn nanocrystals. While nanocrystals may take different forms, our discussions are primarily focused on nanocrystals such as nanowires and nanorods. The design principle and theoretical performance of AlGaIn nanocrystal UV LEDs and surface-emitting UV laser diodes are presented in Section 2. The epitaxy, structural, electronic, and optical properties of AlGaIn nanocrystals are discussed in Section 3. In Section 4, we present recent demonstrations of AlGaIn UV LEDs. The achievement of electrically pumped UV laser diodes with the use of AlGaIn nanocrystals is presented in Section 5. Finally, conclusions are made in Section 6.

2. DESIGN OF AlGaIn NANOCRYSTAL LEDs AND SURFACE-EMITTING LASER DIODES

The light extraction of conventional planar LED structures is often limited by total internal reflection. Such an issue can be potentially addressed by using photonic crystal structures. For example, the incorporation of a periodic photonic crystal structure on the top or bottom of a planar LED structure can effectively couple the guided modes inside the device to radiative modes by diffraction [70–75]. With the use of this approach, a significant light-extraction efficiency ($\sim 73\%$) was demonstrated at ~ 450 nm [76]. A large periodic refractive

index modulation can also be introduced inside an LED structure in the form of a nanowire array or two-dimensional photonic crystal slab. By engineering photonic band structure, e.g., inhibiting the guided mode using a photonic bandgap created by the periodic photonic crystal structure, light emitted from the device can only radiate to ambient medium, thereby leading to a high light-extraction efficiency [77]. The directionality of emitted light may also be controlled using this approach. A potential limitation of this approach, however, is that the internal quantum efficiency may be reduced due to the modification (reduction) of the photonic density of states and radiative recombination at the bandgap. An alternative approach is to enhance light extraction by Bragg scattering while increasing the photonic density of states [33,61,78,79]. In addition, photonic nanocrystals provide the distinct opportunity to engineer laser emission characteristics including threshold current, coupling coefficient, polarization, and far-field emission pattern [80–89].

A. Design of AlGaIn Nanocrystal LEDs

One of the major causes for the extremely low EQE (below 1%) for AlGaIn LEDs in a large part of the deep UV spectrum is the poor LEE. With Al content higher than 70%, photons are mostly emitted in the in-plane direction with TM polarization [27]. As a result, most photons are trapped and reabsorbed inside the device, which not only reduces the LEE but also leads to a severe heating effect. In this section, we discuss the use of photonic nanocrystals to enhance the LEE for such TM-polarized emission. The photonic nanocrystal structure consists of AlGaIn nanocrystals with optimally designed lattice constant and size. Schematically shown in Fig. 1(a), the nanocrystal structure consists of n-Al(Ga)N, a n-AlGaIn cladding layer, a AlGaIn quantum well/dot active region, and a p-AlGaIn layer. Figure 1(b) shows the top view of a photonic crystal structure and illustrates the definition of the lattice constant a and nanocrystal lateral size d . The photonic crystal

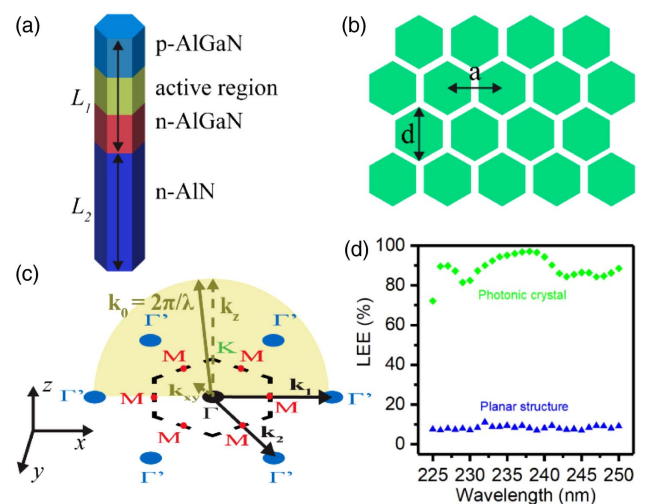


Fig. 1. (a) Schematic of an AlGaIn nanocrystal. (b) Top view of a photonic crystal structure. (c) Schematic for the scattering process in the photonic crystal structure. (d) Light-extraction efficiency (LEE) for the planar structure and the photonic crystal structure with $a = 160$ nm and $d = 95$ nm [61].

structure described here is designed to have a Γ point at around 240 nm. At the Γ point, the in-plane wavevector k_{xy} is negligibly small, as shown in Fig. 1(c). The wavevector is therefore nearly perpendicular to the substrate, indicating that the in-plane emitted light is coupled to the vertical surface emission [90]. By employing UV-reflective substrates such as distributed Bragg reflectors, the light coupled downward can also be reflected and contribute to the device output from the top surface. Through detailed studies using finite-difference time-domain (FDTD) simulation, we have identified that the design with $a = 160$ nm and $d = 95$ nm can exhibit a very large LEE of $>95\%$, which is due to the high out-of-plane coupling coefficient. Moreover, the LEE remains above 80% over a broad wavelength range. For comparison, the LEE of a conventional planar LED structure is also simulated, as shown in Fig. 1(d), which is less than 10%. The LEE is further calculated for practical device configurations that may incorporate a thin p-GaN layer and a thin narrow p-metal grid on top of the nanocrystals, which exhibit an LEE in the range of 30% to 40%. Therefore, the enhancement of LEE by using AlGaN nanocrystals is experimentally feasible.

B. Design of AlGaN Surface-Emitting Laser Diodes

We describe the design and performance characteristics of AlGaN nanocrystal band-edge lasers, which can exhibit surface emission but without the need of the thick and resistive distributed Bragg reflectors required for a conventional VCSEL [62–64,66,67]. By using slow light modes near the Γ point, wherein the in-plane wave vector is nearly zero, photonic crystal structures can be designed to emit light vertically and out of plane, i.e., from the top surface. While light output is designed to be in the vertical direction, there is also lateral leakage from the edge. These two paths of light emission largely determine the Q factor of the lasing mode. Given a certain photonic crystal design, the Q factors associated with vertical and lateral photon confinement generally increase with device area. The Q factors, however, also depend critically on the ratio of nanocrystal size to lattice constant. The maximum coupling to the vertical output and lateral leakage typically occurs at an intermediate ratio of nanocrystal size to lattice constant [91]. Because of the coupling in the vertical direction, some light is also coupled downward to the substrate. In this regard, the use of a reflective substrate/template can readily enhance the useful light output.

Illustrated in Fig. 2(a) (top left inset), AlGaN nanocrystals are arranged in a square lattice with a lattice constant a . AlGaN quantum dots/wells are incorporated in the nanocrystals to serve as the gain medium. Such nanocrystal arrays, with precisely controlled size and spacing, can be achieved using the technique of selective-area epitaxy to be described next [92–95]. To evaluate the device performance, we considered a laser structure consisting of 200 nm n- and p- $\text{Al}_{0.3}\text{Ga}_{0.7}\text{N}$ cladding layers and a 200 nm $\text{Al}_{0.09}\text{Ga}_{0.91}\text{N}$ guiding layer with the incorporation of five GaN (2 nm)/ $\text{Al}_{0.09}\text{Ga}_{0.91}\text{N}$ (4 nm) quantum wells. Shown in Fig. 2(a), the photonic band structure was calculated by the finite-element method (FEM) simulation for nanocrystals with a lateral size of $0.35a$. The simulation was performed using the RF module of Comsol Multiphysics. The refractive index of AlGaN nanocrystals

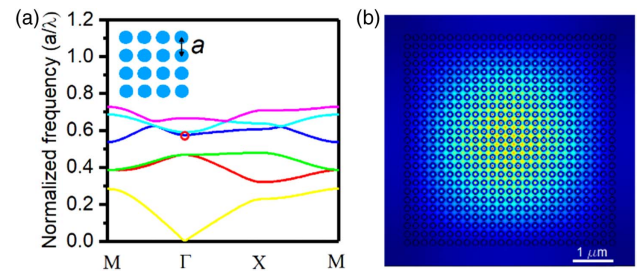


Fig. 2. (a) Photonic band structure of a photonic crystal with a lattice constant of 207 nm and a diameter of 144 nm. The inset is the top view showing the arrangement of nanocrystals. (b) The electric field distribution of the band-edge mode in the entire device.

is 2.65. The normalized frequency of the band-edge mode is 0.575 [shown as the red circle in Fig. 2(a)], which corresponds to $\lambda = 360$ nm for $a = 207$ nm. The group velocity is determined by the slope of the dispersion curve in the photonic band structure. At the band edge, the low group velocity is achieved when the slope of dispersion curve becomes zero ($d\omega/dk \rightarrow 0$), i.e., near the Γ point, the group velocity of light becomes zero, thereby leading to the formation of a stable and large single-cavity mode [96]. Moreover, the extremely low group velocity leads to the long interaction time between radiation field and active material and consequently gives rise to a strong gain enhancement. The intrinsic cavity quality factors are on the order of $(L/\lambda)^3$, where λ is the vacuum wavelength and $L \gg \lambda$ is the lateral dimension of the device structure [96]. Figure 2(b) shows the electric field profile of the band-edge mode calculated with the FEM method. By placing a single dipole source at the center of the simulated structure, the electric field spreads across the entire periodic nanocrystal array, which is a characteristic of the band-edge modes. In this study, photonic crystal arrays in a square region with lateral dimension L in the range of 2.5 to 10 μm are designed and investigated. The relatively large coherent lasing area can lead to excellent beam quality and is well suited for relatively high-output power operation.

1. Threshold Current Density

The performance of a photonic crystal surface-emitting laser can be estimated from a model based on standard rate equations for carriers and photons as follows:

$$\frac{dN}{dt} = \frac{J}{qd} - R_r - R_{nr} - N_{ph}g \frac{c}{n}, \quad (1)$$

$$\frac{dN_{ph}}{dt} = \Gamma_o N_{ph}g \frac{c}{n} - N_{ph}\alpha \frac{c}{n}. \quad (2)$$

The definitions of the parameters are listed in Table 1.

The threshold gain is given by

$$g_{th} = \alpha/\Gamma_o. \quad (3)$$

As the leakage of light at the edge of the device is usually designed to be much smaller than the vertical output, only the loss due to vertical output is considered for calculating the threshold current density. The loss is calculated from the Q factor extracted from 3D FDTD simulation using the relation

Table 1. Definition of Various Parameters Used in the Rate Equations

Parameter	Definition
N	Carrier concentration
N_{ph}	Photon density in the cavity
J	Current density
q	Electron charge
d	Total thickness of the active layers
R_r	Radiative recombination rate
R_{nr}	Nonradiative recombination rate
g	Material gain
c	Light speed in vacuum
n	Refractive index of the active layers
Γ_0	Optical confinement factor
α	Total loss

$$\alpha = \frac{2\pi}{aQ}. \quad (4)$$

The threshold gain is calculated for different device dimensions, and then the threshold current is calculated based on the charge carrier recombination in the active region. Since recombination mostly occurs in the quantum well, any recombination in the bulk region was not taken into account. The nonradiative recombination rate can be written as $R_{\text{nr}} = AN + CN^3$, where A is the Shockley–Read–Hall recombination coefficient and C is the Auger recombination coefficient. To estimate the threshold current, Auger recombination is neglected due to the large bandgap, and A is assumed to be $3 \times 10^8 \text{ s}^{-1}$ [97,98]. The radiative recombination rate R_r at a certain carrier concentration is calculated by integrating the recombination rate over the entire energy distribution of carriers [99]. The optical confinement factor is estimated to be 1.6% from the FEM simulation. The threshold current density is shown in Fig. 3(a). It is seen that the threshold current density decreases drastically with increasing lasing area due to the higher Q . For a lateral size of $10 \mu\text{m} \times 10 \mu\text{m}$, the threshold current density is $\sim 4.9 \text{ kA/cm}^2$ at room temperature. Much smaller threshold current density is expected for devices with a larger Q factor. In practice, the laser threshold depends critically on the design of the active region and the device fabrication process.

2. Lasing Linewidth

Semiconductor lasers with narrow linewidths are very important for emerging quantum applications, including atomic clocks and sensors. To date, the narrowest linewidth of

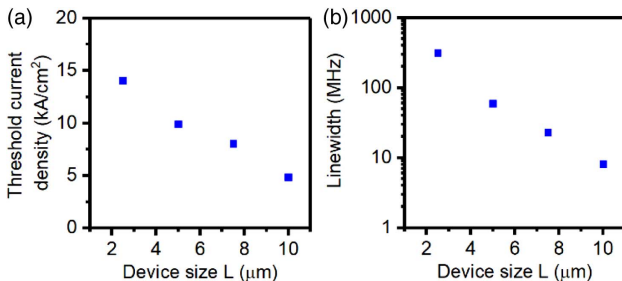


Fig. 3. Variation of (a) threshold current density and (b) linewidth at an injection current of $5 \times I_{\text{th}}$ with the device lateral dimension (L).

VCSELs is $\sim 3 \text{ MHz}$, which was realized by using a very long cavity and by minimizing the mirror loss [100,101]. Such a linewidth, however, is approximately 2 to 3 orders of magnitude larger than the best reported values for distributed feedback (DFB) lasers or distributed Bragg reflector lasers [102,103]. In the spectral range from UV to green, the linewidth of typical diode lasers is on the order of 100 GHz or larger [104–108].

Linewidths of a semiconductor laser are often affected by the Q factor of the cavity mode, output power, and linewidth enhancement factor [7]. The linewidth of a laser can be approximately estimated by [109]

$$\Delta\nu = \frac{R_{\text{sp}}(1 + f_{\alpha}^2)}{4\pi N_{\text{ph}} V_{\text{m}}}, \quad (5)$$

where R_{sp} is the rate of spontaneous emission into the lasing mode, f_{α} is the linewidth enhancement factor, and V_{m} is the mode volume. The photon number is written as [110]

$$N_{\text{ph}} V_{\text{m}} = \frac{1}{\exp[(\hbar\omega - \Delta E_{\text{F}})/kT] - 1}, \quad (6)$$

where $\hbar\omega$ is the photon energy, ΔE_{F} is the difference between quasi-Fermi levels, k is the Boltzmann constant, and T is the temperature. The rate of spontaneous emission into the lasing mode is [110]

$$R_{\text{sp}} = \alpha v_{\text{g}} \exp[(\Delta E_{\text{F}} - \hbar\omega)/kT], \quad (7)$$

where v_{g} is the group velocity. Considering the definitions for α , v_{g} , and Q , we have

$$\alpha = -\frac{dN_{\text{ph}}}{N_{\text{ph}} dx}, \quad (8)$$

$$v_{\text{g}} = dx/dt, \quad (9)$$

$$Q = -\frac{dN_{\text{ph}}}{N_{\text{ph}} dt}. \quad (10)$$

Combining Eqs. (7)–(10), the rate of spontaneous emission into the lasing mode can be written as

$$R_{\text{sp}} = \frac{\omega}{Q[1 + (N_{\text{ph}} V_{\text{m}})^{-1}]}. \quad (11)$$

Because the photon number is typically much larger than one above threshold, the rate of spontaneous emission into the lasing mode is approximately given by

$$R_{\text{sp}} \approx \omega/Q. \quad (12)$$

Above threshold current I_{th} , the photon number at an injection current I can then be expressed as

$$N_{\text{ph}} V_{\text{m}} = \frac{\tau_{\text{ph}}}{q} (I - I_{\text{th}}), \quad (13)$$

where the photon lifetime τ_{ph} can be calculated by

$$\tau_{\text{ph}} = Q/\omega. \quad (14)$$

Based on the analysis above, the linewidth can be written as

$$\Delta\nu = \frac{\omega^2(1 + f_{\alpha}^2)}{4\pi Q^2 q(I - I_{\text{th}})}. \quad (15)$$

In this study, the linewidth was calculated at a current density of $5 \times I_{th}$ for photonic crystal lasers with different lateral dimensions (up to $10 \mu\text{m}$) as shown in Fig. 3(b). A typical linewidth enhancement factor of 2 was used in the calculation [111]. It is seen that the linewidth decreases drastically with increasing device size. This is explained by the larger photon number in the lasing mode and higher Q associated with larger device dimensions. For a photonic nanocrystal surface-emitting laser diode with a lateral dimension $\sim 10 \mu\text{m}$, the linewidth is $\sim 8 \text{ MHz}$, which is 5 to 6 orders smaller than that of previously reported GaN-based VCSELs in the UV-visible spectrum and is on the same order of magnitude of the narrowest linewidths reported for VCSELs operating in the infrared spectrum [100,101,104–108]. The narrowest linewidth of laser diodes reported to date is $\sim 3.6 \text{ kHz}$, which was measured from a DFB laser [103]. It is expected that the linewidth of photonic crystal surface-emitting laser diodes can be potentially reduced to $\sim \text{kilohertz (kHz)}$ by increasing the device size and further optimizing the design.

3. Far-Field Pattern and Mode Profile

Illustrated in Fig. 4 is the simulated far-field pattern using the FDTD method for a photonic crystal structure with a lattice constant of 207 nm and a diameter of 144 nm , showing predominantly a narrow, single-lobed beam due to the coherent oscillation over a large area. Nearly ideal beam quality ($M^2 < 1.1$) and high-output power operation (up to 3.5 W) were achieved previously in InGaAs photonic crystal band-edge lasers operating at 940 nm at room temperature [65]. For the presented photonic nanocrystal surface-emitting laser diodes, by varying the lattice points and/or lattice phases, it is also possible to tune the beam shape and realize surface-emitting laser diodes with on-demand beam characteristics [79,80].

3. EPITAXY AND CHARACTERISTICS OF AlGaN NANOCRYSTALS

Previous studies on the epitaxy of AlGaN nanostructures have been largely focused on spontaneously formed nanowire arrays by using metalorganic vapour-phase epitaxy, or molecular beam epitaxy [29,39–46], which often exhibit random distribution in size, height, and morphology and twist/tilt in crystal orientation [36]. The resultant Al distribution is highly

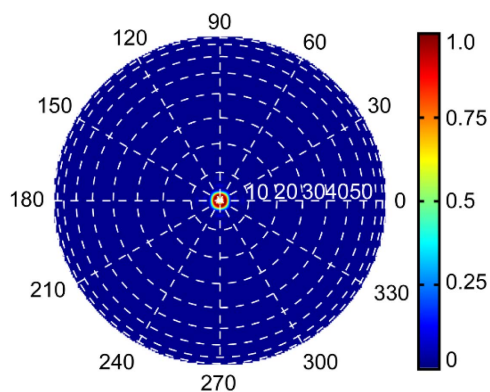


Fig. 4. Far-field radiation pattern calculated for a photonic nanocrystal laser structure with a lattice constant of 207 nm and a diameter of 144 nm .

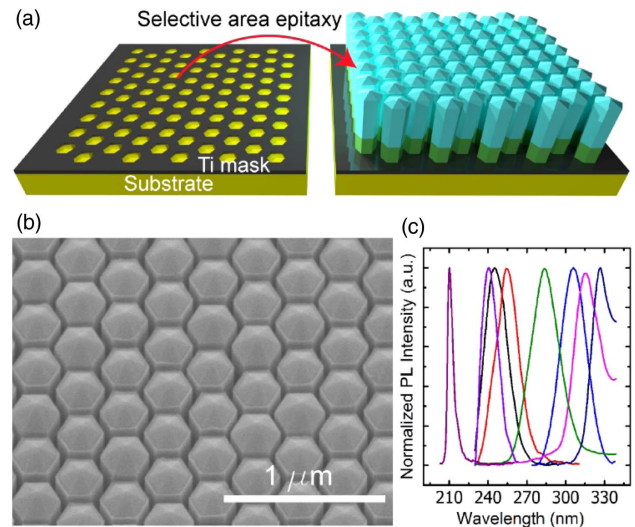


Fig. 5. (a) Schematic of the selective-area epitaxy process. (b) A typical SEM image of AlGaN nanocrystals grown by selective-area epitaxy. (c) Normalized PL spectra for AlGaN nanocrystals with Al content across nearly the entire compositional range [33].

non-uniform, leading to relatively large inhomogeneous broadening [28,29]. In addition, the coalescence of nanowires with misaligned crystal orientation can lead to the formation of extensive defects and dislocations [36]. These issues can be largely addressed by using the technique of selective-area epitaxy [52,53], which can yield AlGaN nanocrystals with precisely controlled size, position, and surface polarity. This growth process is schematically shown in Fig. 5(a). A thin Ti (or SiN_x) layer is first deposited on the substrate surface and patterned by electron beam lithography. Nitridation of the Ti mask is performed at 400°C to prevent Ti cracking during growth [52,55]. GaN (or AlN) nanocrystal array is grown under a relatively high temperature to achieve high growth selectivity. AlGaN is subsequently grown on the GaN (or AlN) nanocrystals. Shown in Fig. 5(b) is the typical scanning electron microscopy (SEM) image of AlGaN nanocrystal array grown by plasma-assisted molecular beam epitaxy (MBE). The Al incorporation can be varied across the entire compositional range by changing the Al beam equivalent pressure during epitaxy [33,112]. Shown in Fig. 5(c) are the photoluminescence (PL) spectra of AlGaN nanocrystals with Al compositions of $\sim 20\%$ to 80% measured at room temperature.

Due to the efficient surface strain relaxation, dislocation-free AlGaN nanocrystals can be grown on foreign substrates. Shown in Fig. 6(a) is $\sim 0.5 \mu\text{m}$ GaN epilayer grown directly on Si(001), which exhibits large densities of pits and dislocations. In contrast, N-polar GaN nanocrystals grown on such GaN/Si templates exhibit smooth surface morphology and are relatively free of pits and dislocations, due to the termination of propagating dislocations on the lateral surfaces of nanocrystals [36]. Moreover, through controlled coalescence process of AlGaN nanocrystals, nearly dislocation-free AlGaN templates can be achieved on sapphire or other foreign substrates [36]. With the incorporation of Mg dopant, the coalesced $\text{Al}_{0.35}\text{Ga}_{0.65}\text{N}$ epilayers can exhibit excellent p-type conduction characteristics,

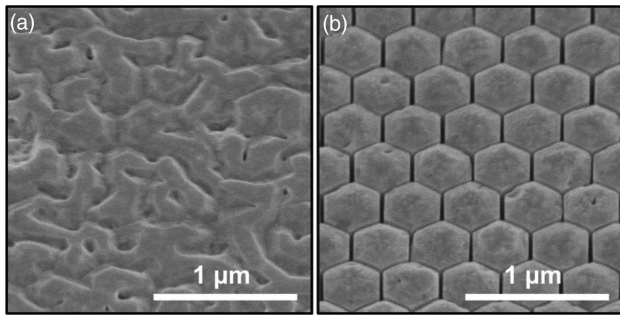


Fig. 6. SEM images of (a) 0.5 μm GaN grown on Si wafer and (b) N-polar GaN nanocrystals grown on Si.

including a free-hole concentration of $\sim 7 \times 10^{18} \text{ cm}^{-3}$ and mobility $\sim 8.85 \text{ cm}^2/(\text{V} \cdot \text{s})$ at room temperature, which are significantly better than previously reported values, respectively [113–115]. High-performance UV LEDs have also been demonstrated with the use of such AlGaIn templates [36]. Significantly, the surface polarity can be controllably tuned to be N-polar, Ga(Al)-polar, or semi-polar when grown on foreign substrates [116–118]. The capacity to realize dislocation-free AlGaIn nanostructures and heterostructures with controlled surface polarity provides a distinct opportunity to significantly enhance the performance of mid- and deep-UV optoelectronic and high-power/frequency electronic devices [33,34,36,42,43,45,119–121].

Dislocation-free AlGaIn nanocrystals also offer unique advantages for achieving efficient p-type conduction that was difficult for conventional AlGaIn epilayers. Shown in Fig. 7(a) are the photoluminescence emission spectra for Mg-doped AlN nanostructures measured at room temperature. The photoluminescence emission of a non-doped sample is also shown for comparison. The pronounced peak at $\sim 230 \text{ nm}$ corresponds to a Mg-acceptor related transition, which is separated from the excitonic emission of AlN by $\sim 0.5\text{--}0.6 \text{ eV}$, in good agreement with previous report for the activation energy for Mg-acceptor level in AlN [122]. Experimentally, we have measured a very small activation energy ($\sim 20\text{--}50 \text{ meV}$ near room temperature) for Mg dopant in AlN nanostructures at high Mg-doping concentrations [49,123]. The hole concentration can reach up to $6 \times 10^{17} \text{ cm}^{-3}$ at room temperature, which is nearly 7 orders of magnitude larger than that of previously reported AlN epilayers [25,49]. Such unusually large carrier concentration is directly related to hole hopping conduction in the Mg

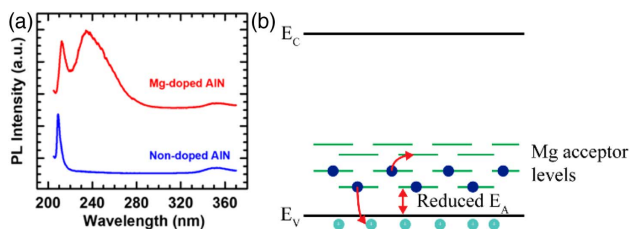


Fig. 7. (a) PL emission spectra of AlN and AlN:Mg nanostructures measured at room temperature. (b) Schematic illustration of the Mg impurity band of AlN nanostructures due to high Mg concentration and the reduced activation energy for a portion of Mg acceptors.

impurity band of AlN nanocrystals. Recent first-principle calculations have shown that Al(Ga) substitutional Mg formation energy is significantly reduced in nanocrystals compared to their planar counterparts [42,124], which can lead to enhanced Mg-dopant incorporation without the formation of extensive defects. The resulting large concentrations of Mg acceptors in Al(Ga)N lead to several critical benefits for efficient p-type conduction. (i) The high Mg acceptor concentration promotes the formation of a Mg impurity band in Al(Ga)N nanocrystals due to the strong interaction of neighboring dopants, which can lead to efficient hole hopping conduction in the impurity band. The two-band conduction model is schematically shown in Fig. 7(b). (ii) In addition, a portion of the Mg dopants have much reduced ionization energy and can be readily ionized and contribute to free holes in valence band due to the band-tailing effect and the broadened Mg energy level distribution, which is evidenced by the partial overlap between the photoluminescence emission of Mg acceptors and the excitonic transition of AlN, as shown in Fig. 7(a). The formation of an impurity band in doped semiconductors has also been reported and analyzed previously [5,42,49,114,125,126]. Also, during the epitaxy of AlGaIn nanocrystals, the growth conditions can be tuned to be N-rich, which makes the formation of N-vacancy-related defects energetically less favorable than metal-rich condition [127]. Furthermore, the luminescence related to such N vacancies was also shown to be absent under N-rich conditions by Shahedipour *et al.*, suggesting the formation of N-vacancy-related defects is suppressed [128]. It is therefore evident that the use of dislocation-free nanocrystals can provide a viable path to achieve efficient p-type conduction in ultra-wide-bandgap AlGaIn that was previously difficult.

4. AlGaIn NANOCRYSTAL UV LEDs

In this section, we review some recent progress on AlGaIn nanocrystal UV LEDs. Due to the enhanced p-type conduction, AlN nanostructured LEDs exhibit excellent current-voltage characteristics. It was demonstrated that these devices could exhibit a turn-on voltage $\sim 6\text{--}7 \text{ V}$, which is much better than that of conventional planar AlN LEDs [25,37,129]. Recently, hexagonal boron nitride (h-BN) was incorporated into AlN LED structures, which led to a significant improvement in device performance [130]. h-BN has been demonstrated to have much higher p-type conductivity than Mg-doped AlN [130]. Recent theoretical calculations showed that certain type of point defects, e.g., boron vacancies, could behave like acceptors [131–133]. In a recent study, h-BN was used to replace conventional p-AlN layer in an AlN LED, shown in the inset of Fig. 8(a). After the growth of AlN nanocrystals, the growth of h-BN was performed at substrate temperatures from 400 to 800°C in a separate MBE system where boron was *in situ* evaporated by electron beam. The growth of BN was conducted under N-rich conditions to promote the formation of boron vacancies, which served as shallow p-type acceptors. The device exhibits excellent current-voltage characteristics with a turn-on voltage of 5.5 V and negligible leakage current under reverse bias. In contrast, the turn-on voltage for conventional AlN epilayer LEDs is larger than 20 V [25]. The electroluminescence (EL) spectra measured under different injection currents at room

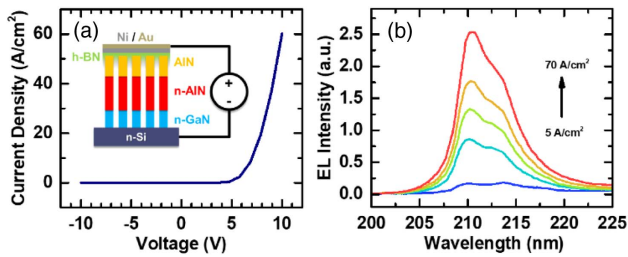


Fig. 8. (a) I - V characteristics of a $300\ \mu\text{m} \times 300\ \mu\text{m}$ LED device at room temperature. Inset: schematic of the fabricated LED structure. (b) Room temperature EL spectra of the LED device for various injection currents [35].

temperature are shown in Fig. 8(b). The main electroluminescence peak is at 210 nm, which is from exciton emission. The presence of a shoulder on the longer wavelength side of the emission spectra is attributed to longitudinal optical phonon replica [134]. The measured light output is more than one order higher than a similar AlN nanowire LED but without the use of h-BN. The improved performance is attributed to more efficient hole transport and injection and better carrier confinement in the active region.

Tunnel injection of nonequilibrium holes has shown to be effective in improving hole injection efficiency and recombination in the active region [32,135–139]. An n^{++} -GaN/Al/ p^{++} -AlGa N tunnel junction was incorporated in AlGa N UV LEDs to improve the hole injection. The EL spectra under different injection currents are shown in Fig. 9(a). A single pronounced peak at 275 nm is measured. Compared to LEDs without a tunnel junction, a drastic improvement in the output power (by nearly 2 orders of magnitude) was measured, as shown in Fig. 9(b). With the use of AlGa N nanostructures, LEDs operating at 240 nm have also been demonstrated with improved performance [140].

5. AlGa N NANOCRYSTAL UV LASER DIODES

Described in Section 2.B, AlGa N nanocrystals offer a distinct opportunity to realize low-threshold UV laser diodes. The first demonstrations of electrically pumped semiconductor laser diodes operating in the UV-B and UV-C bands were realized

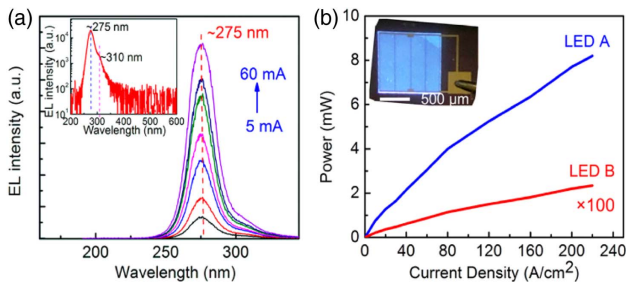


Fig. 9. (a) EL spectra of the Al tunnel junction AlGa N UV LED under CW biasing condition. Inset: EL spectrum in the logarithmic scale. (b) Variations of output power with injection current for Al tunnel junction AlGa N UV LED and standard p-i-n AlGa N UV LED. Inset: an optical image of the device under an injection current of $8\ \text{A}/\text{cm}^2$ [31].

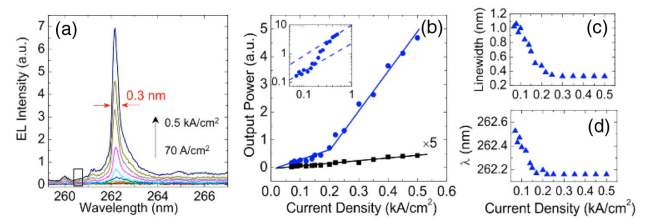


Fig. 10. (a) Emission spectra for an AlGa N laser operating at 262 nm at 77 K under various injection current densities. (b) Variation of output with injection current. Blue circles represent the lasing peak. Black squares represent the background emission in the boxed area in (a) with a linewidth of 0.3 nm. The inset plots the data for the lasing peak in the logarithmic scale. Variations of (c) linewidth and (d) peak wavelength of the lasing peak at 262 nm with injection current density [30].

using AlGa N nanostructures [29,34,141,142]. Such subwavelength nanostructures can form a high- Q optical cavity due to Anderson localization of light [34,143–147]. Shown in Fig. 10(a) are the electroluminescence spectra of a laser diode based on Anderson localization in spontaneously formed AlGa N nanostructures [29]. A sharp and pronounced lasing peak at 262 nm is measured at 77 K. Variations of the light output versus injection current are displayed in Fig. 10(b), showing a threshold current density $\sim 200\ \text{A}/\text{cm}^2$, which is nearly 2 orders lower than recently reported AlGa N quantum well laser diodes at 336 nm and 342 nm [23,148]. The logarithmic plot of the light intensity versus current density is shown in the inset of Fig. 10(b), which exhibits a clear S-shaped trend, further confirming the achievement of lasing. Compared to the lasing emission, the background emission [black squares in Fig. 10(b)] exhibits a small or negligible increase above threshold. The spectral linewidth and the peak wavelength of the lasing mode are plotted in Figs. 10(c) and 10(d), respectively. A significant reduction of the linewidth to $\sim 0.3\ \text{nm}$ was measured at the threshold current density $\sim 200\ \text{A}/\text{cm}^2$, providing unambiguous evidence for the achievement of lasing. Above threshold, the lasing emission peak stays nearly constant at 262.2 nm.

More recently, we have demonstrated an AlGa N laser diode operating at 239 nm at room temperature [142]. Numerical simulation is performed to identify the average diameters and filling factors that can support high- Q random cavity modes around 240 nm. The growth of nanocrystals is optimized for the suited average diameters and filling factors, and the Al content is increased accordingly to tune the emission wavelength to around 240 nm. The emission spectra measured under various injection currents are shown in Fig. 11(a), which are characterized by broad spontaneous emission below threshold and a sharp lasing peak above threshold. Variations of the light output for the lasing mode versus injection current are shown in Fig. 11(b), which exhibits a distinct threshold current $\sim 0.35\ \text{mA}$. The light intensity versus injection current exhibits a clear S shape when plotted in a logarithmic scale, shown in the inset of Fig. 11(b), further confirming the achievement of lasing. For comparison, the background emission around 267 nm indicated by the black box in Fig. 11(a) was also analyzed,

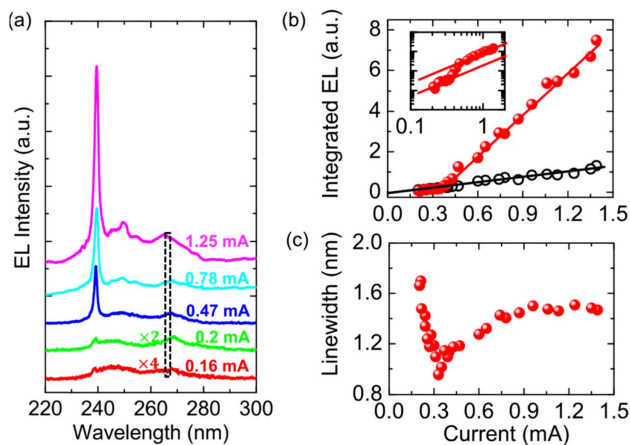


Fig. 11. (a) Emission spectra of an AlGaIn laser device operating at 239 nm in CW biasing condition under different injection currents. (b) Variation of output with injection current for the lasing peak (red filled circles) and a non-lasing cavity mode (black open circles) from the boxed region in (a). The inset plots the data for the lasing peak in the logarithmic scale. (c) Variations of linewidth with the injection current [142].

which only exhibited a very small increase with current above threshold [black circles in Fig. 11(b)]. Variations of the spectral linewidth of the lasing peak versus injection current are shown in Fig. 11(c). A clear reduction of the linewidth is seen at threshold.

6. CONCLUSION

In summary, dislocation-free AlGaIn nanocrystals hold tremendous promise to break the efficiency bottleneck of UV optoelectronics. With the use of selective-area epitaxy by MBE, their structural, electronic, and optical properties can be controlled. Through photonic band gap engineering, AlGaIn nanocrystal LEDs promise significantly improved light-extraction efficiency for TM-polarized photons. AlGaIn nanocrystals also provide a viable path to achieve electrically pumped semiconductor lasers in the UV-B and UV-C bands, including both edge- and surface-emitting devices with superior performance. The current challenges include the development of UV-transparent electrode and passivation materials, which are essentially required to achieve efficient AlGaIn nanocrystal LEDs and laser diodes in the mid- and deep-UV spectra.

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