Supporting Information

Engineering the Carrier Dynamics of InGaN Nanowire White Light-Emitting Diodes by Distributed *p*-AlGaN Electron Blocking Layers

Hieu Pham Trung Nguyen[1,2], Mehrdad Djavid[1], Steffi Y. Woo[3], Xianhe Liu[1], Ashfiqua. T. Connie[1], Sharif Md.Sadaf[1], Qi Wang[1], Gianluigi A. Botton[3], Ishiang Shih[1], and Zetian Mi[1*]

[1]*Department of Electrical and Computer Engineering, McGill University*

3480 University Street, Montreal, Quebec H3A 0E9, Canada

[2]*Department of Electrical and Computer Engineering, New Jersey Institute of Technology, University Heights, Newark, New Jersey 07102*

[3]*Department of Materials Science and Engineering, Canadian Centre for Electron Microscopy and Brockhouse Institute for Materials Research,*

McMaster University, 1280 Main Street West, Hamilton, Ontario L8S 4L7 Canada

[*]*E-mail: zetian.mi@mcgill.ca*

S1. Optical Characterization

Photoluminescence (PL) spectra of the InGaN/AlGaN dot-in-a-wire core-shell LEDs measured using a 266 nm laser at room-temperature are shown in Figure S1. It is seen that the PL intensity, for both the InGaN quantum dot active region and GaN, increases dramatically with the presence of AlGaN shell, compared to the InGaN/GaN dot-in-a-wire structure. Moreover, the PL intensity shows an increasing trend with increasing Al compositions in the AlGaN barrier layers for LED 1, LED 2, and LED 3. For LED 4, the PL intensity shows a decrease, possibly due to the presence of structural defects in the device active region with very high Al composition.

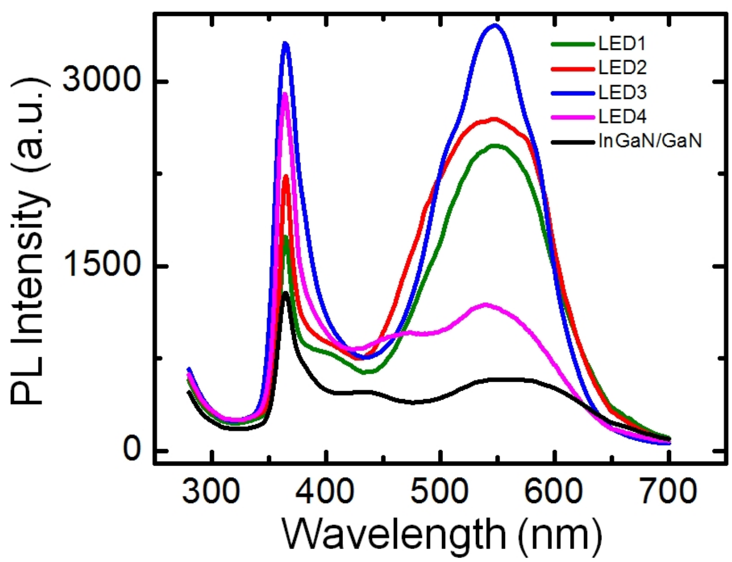


Figure S1: Photoluminescence spectra of the InGaN/AlGaN dot-in-a-wire core-shell LED structures, including LED 1, LED 2, LED 3, and LED 4. The photoluminescence spectrum of the InGaN/GaN dot-in-a-wire sample without the presence of AlGaN shell is also shown for comparison.

Moreover, PL spectra of the nanowire LEDs were also measured using a 405 nm laser as the excitation source at room-temperature. Shown in Figure S2, the PL intensity of the

InGaN/AlGaN dot-in-a-wire core-shell sample (LED 3) is more than 7 times higher than that of the InGaN/GaN dot-in-a-wire LED structure without the presence of AlGaN shell. Given the nearly identical nanowire size, density, and morphology, the significantly enhanced PL intensity of InGaN/AlGaN dot-in-a-wire core-shell LED structures is attributed to the reduced nonradiative surface recombination, due to the superior carrier confinement offered by the large bandgap AlGaN shell.

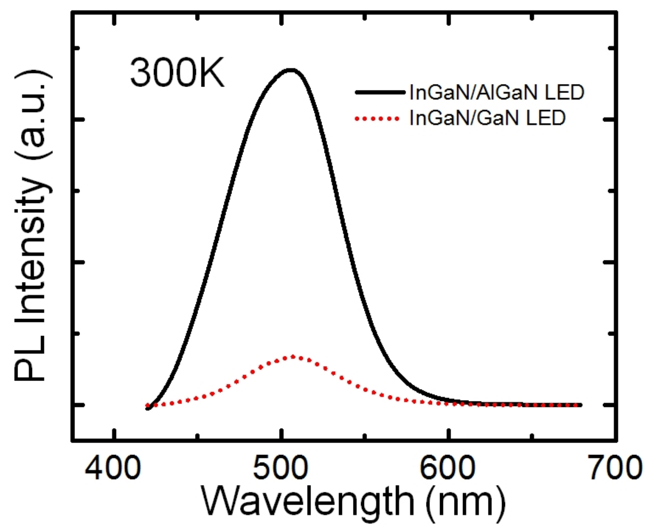


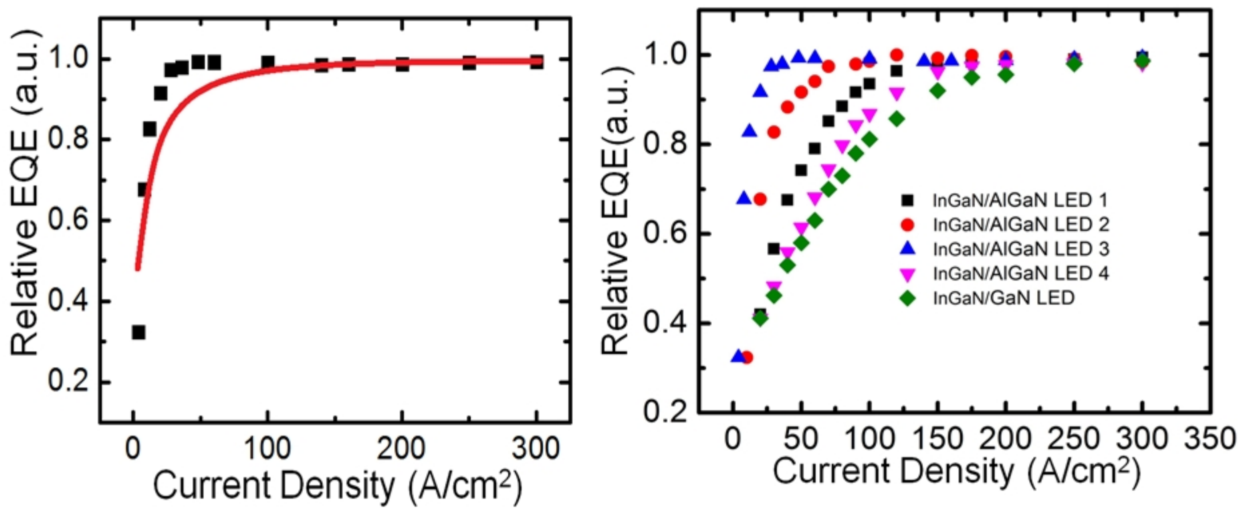
Figure S2: Photoluminescence spectra of InGaN/AlGaN dot-in-a-wire core-shell LED structure (LED 3) and the InGaN/GaN dot-in-a-wire structure without AlGaN shell measured using a 405 nm laser at room-temperature.

S2. Electroluminescence Characterization

The current-voltage characteristics of the nanowire LEDs were measured using a pulse generator (Avtech AV-1010-B). Electroluminescence emission of the LED devices were collected by an optical fiber and analyzed using an Ocean optics spectrometer. Additionally, we have calculated the relative external quantum efficiency (EQE) of the InGaN/AlGaN dot-in-a-wire core-shell LED devices, which is derived by dividing the integrated electroluminescence intensity by the corresponding injection current. Shown in Figure S3(a) for LED 3 with device size of ~500μm×500μm, the room temperature EQE of such core-shell nanowire LEDs exhibits a fast rising trend. The peak EQE is measured at relatively low injection current (~ 50 A/cm^2), which is significantly lower than that of conventional InGaN/GaN axial nanowire LEDs (generally higher than 200 A/cm^2)[1-3]. We have further analyzed the relative EQE of the core-shell nanowire LEDs using the ABF model, described below,

$$\eta_i = \frac{BN^2}{AN+BN^2+f(N)} \quad (1)$$

where η_i is the device internal quantum efficiency, and N is the carrier density in the device active region. A and B are the Shockley-Read-Hall (SRH) nonradiative recombination coefficient and the radiative recombination coefficient, respectively. $f(N)$ represents for the Auger recombination, electron overflow and/or other higher order carrier-loss processes. Illustrated in Figure S3(a), the derived A coefficient is ~ 1×10^7 s^{-1}, which is significantly lower than the previously reported values of InGaN/GaN nanowire LEDs without using AlGaN shell[1]. This study further confirms the positive effect of AlGaN shell on the reduced nonradiative surface recombination, which is also consistent with the PL and carrier lifetime results shown in the main text. The B coefficient is estimated to be ~ 1.75×10^{-9} cm^3 s^{-1}, which is higher than that of nanowire LED without using core-shell structure[2]. The Auger coefficient is very low, which is ~ 1×10^{-33} cm^6s^{-1}, suggesting that Auger nonradiative recombination and/or electron overflow has a negligible impact on the device performance. Figure S3(b) shows the relative EQEs of LED 1, LED 2, LED 3, LED 4 and InGaN/GaN LED versus injection current. A fast rising trend was observed for nanowire LEDs using AlGaN shells compared to InGaN/GaN LEDs. Moreover, peak EQEs are recorded at much lower injection currents compared to InGaN/GaN LEDs without using AlGaN shell, further confirming the reduced nonradiative surface recombination in such InGaN/AlGaN core-shell LEDs.



(a) (b)

Figure S3: (a) Relative external quantum efficiency (EQE) of an InGaN/AlGaN dot-in-a-wire core-shell LED 3 with device size of 500μm×500μm measured at room-temperature. Also shown in the figure is the simulated internal quantum efficiency (solid red curve) based on the ABF model. (b) Relative EQE of InGaN/AlGaN core-shell LEDs and InGaN/GaN LEDs versus injection currents.

S3. Device Modeling

The optical and electrical properties of InGaN/AlGaN LEDs, including the output power and the electron and hole distributions are numerically calculated using the APSYS simulation software and compared to those of InGaN/GaN LED structures with and without the presence of an equivalent electron blocking layer (EBL). Schematics of the simulated structures are shown in Figure S4. The APSYS is based on 2D finite element analysis of electrical and optical properties of semiconductor devices solving the Poisson's equation, carrier transport equation, and photon rate equation. Shown in Figure S4, the device active region consists of InGaN (3 nm)/(Al)GaN (3 nm) superlattice structures. An average In composition of 20% is used. For the core-shell structure, the Al composition in the AlGaN barrier layer is ~ 20%, shown in Figure S4(c). An equivalent AlGaN EBL (thickness ~ 30 nm) is also incorporated in the InGaN/GaN LED structure, illustrated in Figure S4(b). In this study, 20% and 15% of the theoretical polarization induced sheet charge density is used for InGaN/AlGaN and InGaN/GaN LED structures, respectively. In this two-dimensional simulation, Shockley−Read−Hall lifetime and Auger coefficient are assumed to be 4.5 ns and 1×10^{-34} cm^6s^{-1}, respectively. Nonradiative surface recombination was not considered.

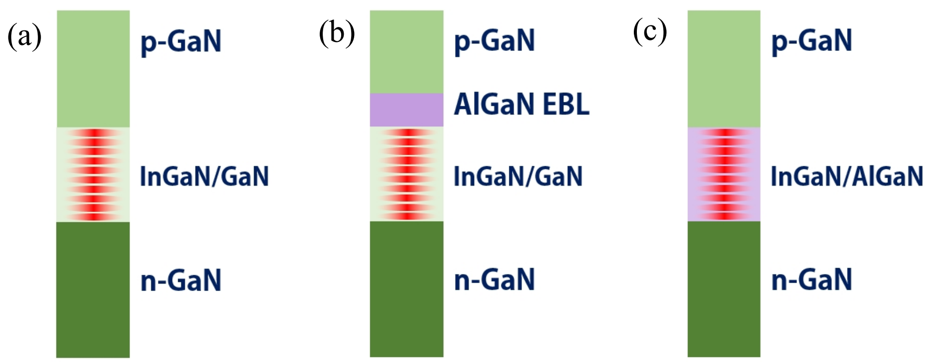


Figure S4: Schematics of (a) the InGaN/GaN LED heterostructure, (b) the InGaN/GaN LED with an equivalent AlGaN EBL, and (c) the InGaN/AlGaN LED heterostructure.

The use of AlGaN EBL has been previously proposed and implemented to overcome the leakage of electrons outside of the LED active region[2,4-6]. However, in order to form a relatively thick AlGaN shell surrounding the device active region, a relatively thick (~ 30 nm) AlGaN EBL is required for the nanowire LEDs, which adversely affects the hole transport and injection into the active region, leading to highly nonuniform carrier distribution[5,7-9]. We show that such an issue can be largely addressed by using distributed AlGaN EBL, i.e. InGaN/AlGaN superlattice structures. The presence of multiple AlGaN barrier layers in the device active region can lead to significantly reduced electron leakage. Moreover, illustrated in Figures S5, detailed simulation shows that the InGaN/AlGaN LEDs can exhibit more uniform electron and hole concentrations and significantly enhanced power, compared to InGaN/GaN LEDs. The improvement in efficiency droop is also evident in Figure S5(c).

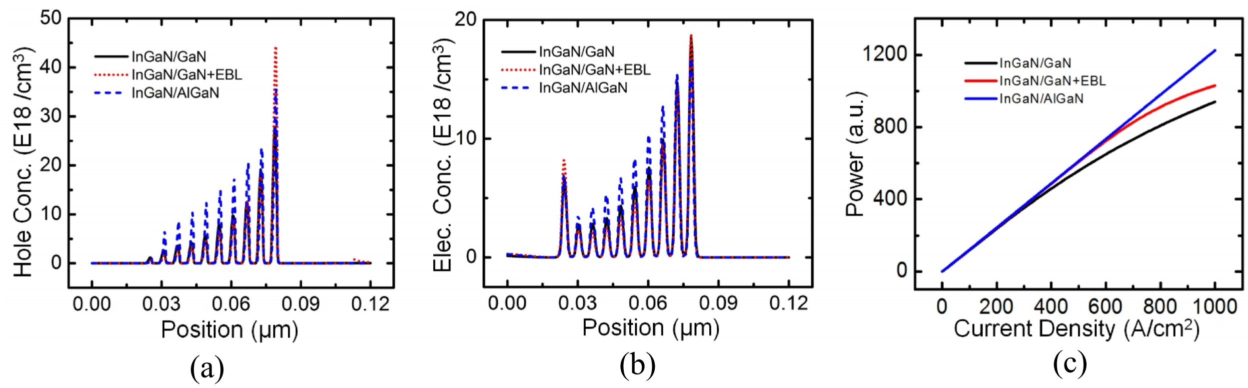


Figure S5: Simulated hole concentration (a) and electron concentration (b) in the active regions of the InGaN/AlGaN (dashed blue), the InGaN/GaN with an equivalent EBL (dotted red), and the InGaN/GaN LED heterostructures (black). (c) Simulated output power vs. injection current for the three LED structures. In this simulation, the effect of nonradiative surface recombination was not considered, which can lead to dramatically reduced power for conventional InGaN/GaN axial nanowire LEDs.

References:

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