Applied Physics Letters

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Citation: Appl. Phys. Lett. **98**, 193102 (2011); doi: 10.1063/1.3588201
View online: http://dx.doi.org/10.1063/1.3588201
View Table of Contents: http://apl.aip.org/resource/1/APPLAB/v98/i19
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InGaN/GaN disk-in-nanowire white light emitting diodes on (001) silicon

Wei Guo,[1] Animesh Banerjee,[1] Pallab Bhattacharya,[1,a)] and Boon S. Ooi[2]
[1]*Department of Electrical Engineering and Computer Science, Center for Nanoscale Photonics and Spintronics, University of Michigan, Ann Arbor, Michigan 48109-2122, USA*
[2]*Division of Physical Sciences and Engineering, King Abdullah University of Science and Engineering, Thuwal 23955-6900, Saudi Arabia*

(Received 10 March 2011; accepted 14 April 2011; published online 9 May 2011)

High density ($\sim 10^{11}$ cm^{-2}) GaN nanowires and InGaN/GaN disk-in-nanowire heterostructures have been grown on (001) silicon substrates by plasma-assisted molecular beam epitaxy. The nanowires exhibit excellent uniformity in length and diameter and a broad emission is obtained by incorporating InGaN disks of varying composition along the length of the nanowires. Monolithic lighting emitting diodes were fabricated with appropriate n- and p-doping of contact layers. White light emission with chromaticity coordinates of $x=0.29$ and $y=0.37$ and a correlated color temperature of 5500–6500 K at an injection current of 50 A/cm^2 is measured. The measured external quantum efficiency of the devices do not exhibit any rollover (droop) up to an injection current density of 400 A/cm^2. © *2011 American Institute of Physics*. [doi:10.1063/1.3588201]

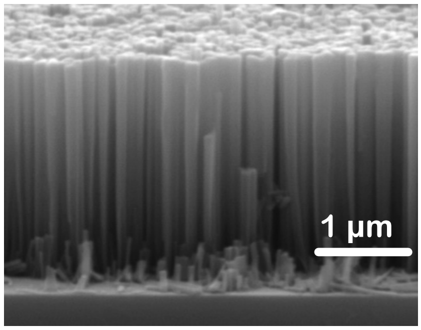
There is a need to develop phosphor-free solid state white light sources. This can be achieved with nitride-based light emitting diodes (LEDs) in which InGaN/GaN quantum wells in the active region are tuned to obtain either a broad emission or emission of dual or multiple wavelengths, which combine to produce white light.[1–6] A shortcoming associated with the used of quantum wells is the large quantum confined Stack effect (QCSE) in the wells and the resulting blueshift in emission wavelength with applied bias.[6] It has been shown that the wavelength shift due to QCSE, arising from the large polarization field in quantum wells,[7] is absent in nanorod LEDs due to the absence of such fields.[8,9] The use of nanowires in the design of white LEDs is important and advantageous for a number of reasons: Ga(In)N nanowires can be grown directly, using Ga as a self-catalyst,[10] on (001) Si (Ref. 8) which is technologically important; extensive structural characterization by several groups have indicated that the nanowires are free of extended defects such as dislocations, stacking faults, and twins;[11–16] the surface recombination velocity on GaN nanowires are 2 orders of magnitude smaller than that on the free surface of GaAs;[17] the Auger coefficients measured in In(Ga)N nanowires and InGaN/GaN disk-in-nanowire are 2–3 orders of magnitude smaller than those measured in heteroepitaxial bulk materials with defects present.[18] This is important in the context of device efficiency. In the present study we have measured the characteristics of monolithic InGaN/GaN disk-in-nanowire white LEDs grown on (001) Si substrates.[8] The composition of the disks has been progressively tuned to obtain multiwavelength luminescence and white light emission. No rollover, or droop, is observed in the measured external quantum efficiency (EQE), up to an injection current density of 400 A/cm^2. Correlated color temperatures (CCT) of 5500–6500 K are derived from the Planckian locus.

GaN nanowires with a density of $\sim 1\times 10^{11}$ cm^{-2} were grown on (001) Si substrate with resistivity <0.001 Ω cm in a Veeco plasma-assisted molecular beam epitaxy system. After removal of the surface oxide on the substrate with a 900 °C anneal in the growth chamber, the substrate temperature is lowered to 800 °C and a few monolayers of Ga are deposited with a Ga flux of 1.5×10^{-7} Torr in the absence of N. GaN nanowire growth is initiated at the same temperature at a rate of 300 nm/h under N-rich conditions. The Ga flux is maintained at 1.5×10^{-7} Torr and the N flow rate is held constant at 1 SCCM (SCCM denotes cubic centimeter per minute at STP). InGaN and GaN layers are grown alternatively to form nanowires containing InGaN/GaN disk-in-nanowire heterostructures. To grow InGaN quantum disks with different In compositions, the Ga and In fluxes are held constant at 1×10^{-7} Torr and 1.5×10^{-7} Torr, respectively, and the growth temperature is varied from 500 to 580 °C.

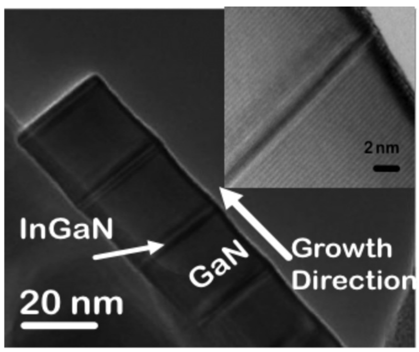
The structural properties of the nanowires were investigated by scanning electron microscope and high resolution transmission electron microscope (HR-TEM) imaging. As shown in Fig. 1(a), high density ($\sim 10^{11}$ cm^{-2}) GaN nanowires are grown with diameters ranging from 10 to 50 nm (in different samples) and they exhibit excellent uniformity in length. The TEM image of Fig. 1(b) shows a GaN nanowire with disk-in-nanowire heterostructure, where multiple layers of InGaN disks of 2 nm thickness were self aligned along the nanowire growth direction. The inset high resolution TEM image depicts a smooth and dislocation-free interface between InGaN and GaN. It is known from selective area diffraction measurements that the nanowires grow in the wurtzite crystalline structure with the c-axis parallel to the direction of growth.[8] Due to the relatively large diameter of the base of the quantum disks, the quantum confinement in the InGaN disks is primarily provided in the growth direction. It is also of interest to note that the size of the InGaN regions, shaped as disks, is almost identical to that of self-assembled InGaN/GaN quantum dots.[19]

The optical properties of the nanowires were examined by photoluminescence (PL) measurements. As illustrated in Fig. 2(a), room temperature PL spectra with peak wavelengths ranging from blue to red were obtained from disk-in-nanowire heterostructures with different indium compositions in the InGaN disks. The dotted curve in Fig. 2(b) represents the PL spectrum obtained from a 300 nm InGaN

a)Electronic mail: pkb@eecs.umich.edu. Tel.: 734-763-6678. FAX: 734-763-9324.

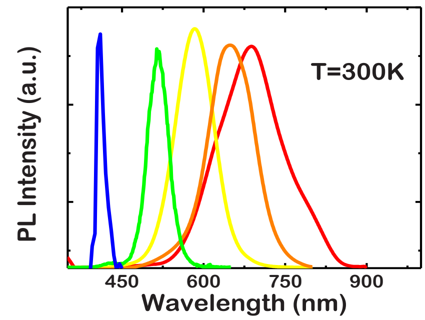


(a)

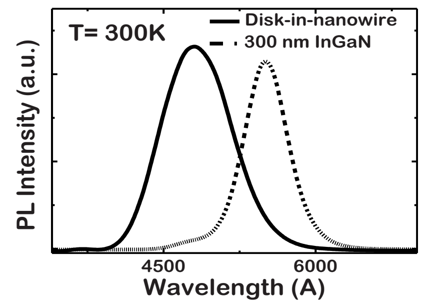


(b)

FIG. 1. (a) High density ($\sim 10^{11}$ cm^{-2}) GaN nanowires grown on (001) Si and (b) HR-TEM image showing a GaN nanowire with multiple InGaN/GaN disk-in-nanowire heterostructures. The high resolution cross-TEM image shown in the inset depicts a single disk-in-nanowire.



(a)



(b)

FIG. 2. (Color online) (a) Room temperature PL spectra of different InGaN/GaN disk-in-nanowire samples illustrating the tunability in emission wavelength obtained with change in InGaN disk alloy composition and (b) PL spectra obtained from InGaN/GaN disk-in-nanowire heterostructure (solid line) and 300 nm InGaN nanowire with similar dimension (dotted line) with identical alloy composition. The disk-in-nanowire sample exhibits a blueshift in 50 nm mainly due to quantum confinement.

nanowire with dimensions similar to a nanowire with quantum disks (disk-in-nanowire structure). The growth of such InGaN nanowires has been described in an earlier publication.[8] In comparison, the solid curve in Fig. 2(b) shows the PL spectrum of a InGaN/GaN disk-in-nanowire heterostructure having 10 InGaN quantum disks (2 nm) separated by 20 nm GaN barriers. The disks have the same indium composition as in the InGaN nanowire. A 50 nm blueshift is observed in the peak emission of the InGaN/GaN disk-in-nanowire structure with respect to the spectrum of the InGaN nanowires, which is largely due to quantum confinement along the growth direction in the InGaN/GaN quantum disks.

LEDs were fabricated with p-i-n InGaN/GaN disk-in-nanowire heterostructures in which the composition of successive InGaN disks along the length of the nanowire is varied. The diode samples were grown on n-type (001) Si substrate. 300 nm of Si-doped GaN nanowire is first grown, followed by the active region consisting of 12 stacks of 2 nm InGaN disk/20 nm GaN barrier layer. The diameter of the InGaN disks is the same as that of the GaN nanowires with an average value of ~30 nm. In order to achieve "white" emission, the 12-period InGaN/GaN disks were divided into three groups. Each group of 4-period InGaN/GaN disks was grown at a different temperature, ranging from 500 to 580 °C. Finally, 150 nm Mg-doped p-type GaN was grown on top. The nanowires were planarized with a parylene insulating layer and covered with 5 nm/5 nm Ni/Au and 250 nm indium tin oxide (ITO) as the top Ohmic contact to the p-GaN nanowires. Aluminum was deposited on the n-type Si to form the bottom electrode. Both parylene and ITO are nearly transparent to visible light. A comment should be made regarding the role of parylene, which is used for planarization in device processing. It is evident, from time-resolved PL measurements made on bare nanowires and those embedded in parylene, that the polymer serves to passivate surface states and reduce surface depletion.[20] Specifically, the relaxation time (~500 ps) of a defect-bound exciton, believed to originate from surface states of the nanowires, decreases and approaches the relaxation time constant of free excitons (~200 ps) from the bulk of the nanowires after coating the nanowires with parylene. This decrease is explained by considering a reduction in surface state density and the associated surface depletion layer width, resulting in a higher degree of electron-hole overlap and a shorter lifetime.

Figure 3(a) depicts the measured room temperature current-voltage (I-V) characteristics of a typical disk-in-nanowire diode, which is schematically shown in this figure. A turn-on voltage of 5 V and series resistance of 18 Ω is measured. The inset of Fig. 3(a) shows the electroluminescence spectra, under continuous wave (cw) bias, of a InGaN/GaN disk-in-nanowire white LED with the injection current density varied from 20 to 50 A/cm^2. The mesa size is 900 $\times 900$ μm and the filling factor of the nanowires is estimated to be ~35%. Light-current characteristics were also measured under pulsed bias conditions (pulse width 1 ms, duty cycle 0.1%) to minimize Joule heating effects. The duty cycle was varied till two sets of data coincided, confirming that such effects are minimized. Measured characteristics are shown in Fig. 3(b). The corresponding EQE in arbitrary units is also plotted in Fig. 3(b). It is evident that no efficiency

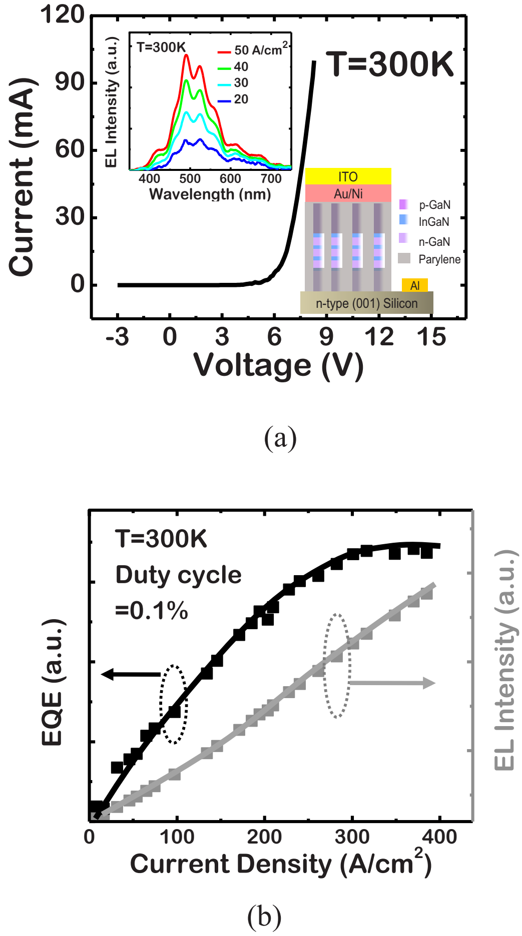


FIG. 3. (Color online) (a) Measured room temperature I-V characteristics of a typical nanowire LED. The inset shows the electroluminescence spectra, under cw bias condition, of a multidisk layer white emission InGaN/GaN nanowire LED with the injection current density varied from 20 to 50 A/cm^2; and (b) plots of light intensity and EQE in arbitrary units vs injection current density.

droop occurs up to $\sim 400\ A/cm^2$. The Commission Internationale de I'Eclairage chromaticity coordinates of $x=0.29$ and $y=0.37$ are derived by analyzing the electroluminescence spectrum of the LEDs under a forward-bias current density of 50 A/cm^2. CCT of 5500–6500 K are also derived from the Planckian locus.

The excitation dependent electroluminescence data shown in the inset of Fig. 3(a) agree with earlier reports on similar measurement on bulk nanowires. While the piezoelectric field may be very small in the nanowires, a finite spontaneous polarization field is present. Our data suggests that a compensating effect, such as Coulomb renormalization,[21] is present to produce negligible shift in the emission with increasing excitation. From the measurement of Auger recombination in InGaN nanowires and InGaN/GaN disk-in-nanowires, we had found that the Auger coefficient C_0 is $\sim 10^{-33}$–10^{-34} $cm^6\ s^{-1}$.[18] These values are ~ 3 orders of magnitude smaller than those measured in InGaN/GaN quantum wells and quantum dots and are in agreement with theoretically calculated ones.[22–26] We, therefore, attribute the absence of droop in the efficiency characteristics of the nanowire LEDs to the absence of any significant Auger recombination. We have not analyzed the efficiency data in an attempt to determine the A-B-C coefficients since multiple wavelengths are present in the emission spectra. Finally, a comment on the nature of white light observed in the nanowire LEDs should be made. The white light emitted by the LEDs reported here would be described as "cool white." In order to get "warm white" emission, the luminescence spectrum shown in the inset of Fig. 3(a) will have to be shifted to longer wavelengths. This can be achieved by increasing the indium composition in the InGaN disks in each group of disks in the active region.

In conclusion, we report the characteristics of monolithic InGaN/GaN disk-in-nanowire LEDs grown on (001) silicon substrate. White light emission with a CC$\sim$5500–6500 K was achieved by tuning the alloy composition of successive disks along the nanowire. No efficiency rollover is observed in the output characteristics.

The work is supported by Grant No. N012509-00 from KAUST, Saudi Arabia.

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