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# Nonradiative Recombination Mechanism in Phosphor-Free GaN-Based Nanowire White Light Emitting Diodes and the Effect of Ammonium Sulfide Surface Passivation

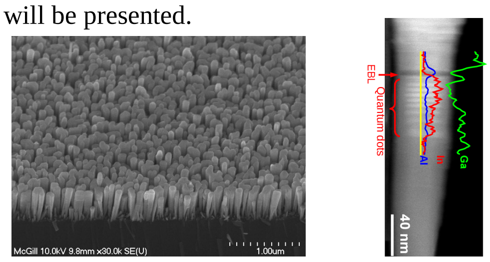
**H. P. T. Nguyen, M. Dajvid, and Z. Mi***
*Department of Electrical and Computer Engineering, McGill University, 3480 University Street, Montreal, Canada H3A 2A7*
*: Phone: 514 398 7114; E-mail: zetian.mi@mcgill.ca*

III-nitride nanowire structures have been intensively studied and shown tremendous promise for applications in solid state lighting. Compared to their conventional planar counterparts, III-nitride nanowires can exhibit greatly reduced dislocation densities and polarization fields, due to the effective lateral stress relaxation. Additionally, the incorporation of quantum dots in the nanowire LED active region may effectively provide superior carrier confinement, promising high efficiency LEDs with tunable emission [1]. To date, however, the quantum efficiency of nanowire LEDs generally exhibits a very slow rise with injection current, indicating the presence of significant nonradiative carrier recombination processes. Through detailed temperature-dependent electroluminescence and simulation studies of GaN-based nanowire white LEDs, we have found that nonradiative recombination at the nanowire surface plays a dominant role in limiting the nanowire LED performance [2]. We have further shown that the quantum efficiency at low injection current can be significantly enhanced by employing a suitable ammonium sulfide passivation technique, due to the greatly reduce nonradiative surface recombination.

Catalyst-free InGaN/GaN dot-in-a-wire nanoscale LED heterostructures are grown by RF plasma-assisted molecular beam epitaxy on Si(111). We have recently demonstrated ultrahigh efficiency intrinsic white light emission by varying the sizes and/or compositions of the dots. The LED active region consists of 10 vertically aligned InGaN/GaN quantum dots, and a p-doped AlGaN electron blocking layer (EBL) is incorporated between the quantum dot active region and the p-GaN to reduce the electron overflow. Shown in figure 1(a) is the scanning electron microcopy (SEM) image of a typical nanowire LED sample. It is seen that the nanowires are highly uniform and vertically aligned to the Si substrate. Ten InGaN/GaN dots and an AlGaN EBL can also be clearly identified in the TEM image shown in figure 1(b). The nanowire LED fabrication process includes standard photolithography, surface passivation with polyimide, and contact metallization techniques. LED devices with areal size of 300×300 $\mu m^2$ were used for detailed characterization.

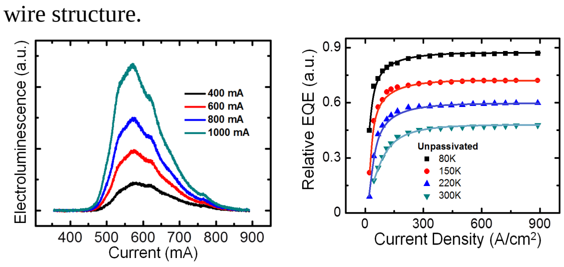
Electroluminescence and quantum efficiency of these nanowire LEDs were measured under pulsed biasing conditions (duty cycle of ~ 0.1%) to effectively reduce junction heating effect. Illustrated in figure 2(a), strong white light emission with broad spectral linewidths (full-width-half-maximum ~ 155nm) was measured, which is attributed to the compositional/size variations of the dots in the active region. Figure 2(b) shows the relative external quantum efficiency of an InGaN/GaN dot-in-a-wire LED measured at various temperatures. It is seen that the nominal maximum EQE is first reached at relatively high injection current of ~ 150A/cm$^2$ at 80K and at ~ 250A/cm$^2$ at 300K. Such values are much higher compared to those of conventional InGaN/GaN quantum well blue LEDs, which are generally reported in the range of ~ 10 to 20A/cm$^2$. We have further calculated the nonradiative recombination coefficient (A) by analyzing the internal quantum efficiency of nanowire LEDs using the ABF model, which are estimated to be in the range of ~ $10^8 s^{-1}$ at room temperature. The large values of A can be well explained by the nonradiative recombination related to the surface states/defects. In nanowire devices, the presence of surface states and defects, due to the large surface-to-volume ratios, can contribute significantly to the carrier loss. However, surface states/defects of nanowire LEDs can be effectively passivated by using ammonium sulfide solution, which can lead to drastically reduced surface recombination velocity. We have studied the fabrication and performance characteristics of such passivated LED devices. Shown in figure 3(a), in the temperature range of 80 to 300K, the injection currents at which the quantum efficiency reaches ~ 80% of its peak value are measured to be in range of 50 to 200A/cm$^2$ and 35 to 61A/cm$^2$ for the unpassivated and passivated devices, respectively. The derived A coefficient varies from ~ $8.4\times10^7$ to $1.83\times10^8 s^{-1}$ from 80K to 300K for the unpassivated LED and is greatly reduced to the range of $0.78\times10^7$ to $4.63\times10^7 s^{-1}$ for the passivated LED. The significantly reduced A values are attributed to the much reduced nonradiative recombination at the nanowire surface due to the effect of ammonium sulfide passivation technique [3,4].

The achievement of high power, highly reliable dot-in-a-wire white LEDs is being investigated and will be presented.



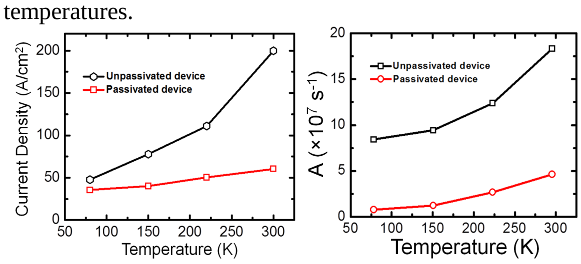

(a) (b)

**Figure 1:** (a) Scanning electron microscopy image of a typical dot-in-a-wire LED sample. (b) Bright field TEM image and EDXS analysis of a single InGaN/GaN dot-in-a-wire structure.




(a) (b)

**Figure 2:** (a) Room-temperature electroluminescence spectra of a nanowire white LED. (b) Variations of the relative EQE with injection current measured at different temperatures.




(a) (b)

**Figure 3:** (a) Injection current density at which the relative EQE reaches its 80% peak value vs. temperature. (b) Derived nonradiative recombination A vs. temperature for both unpassivated and passivated LED devices.

[1] H. P. T. Nguyen *et al.*, *Nano Lett.*, **11**, 1919 (2011). [2] H. P. T. Nguyen *et al.*, *Nanotechnology*, **23,** 194012 (2012). [3] Y. H. Chang *et al.*, *J. Appl. Phys.*, **107**, 043710 (2010). [4] G. L. Martinez *et al.*, *J. Electron. Mater.*, **29,** 325 (2000).