Fabrication of Phosphor-Free III-Nitride Nanowire Light-Emitting Diodes on Metal Substrates for Flexible Photonics

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ABSTRACT: In this paper, we report our study on high-performance III-nitride nanowire light-emitting diodes (LEDs) on copper (Cu) substrates via the substrate-transfer process. Nanowire LED structures were first grown on silicon-on-insulator (SOI) substrates by molecular beam epitaxy. Subsequently, the SOI substrate was removed by combining dry- and wet-etching processes. Compared to conventional nanowire LEDs on Si, the nanowire LEDs on Cu exhibit several advantages, including more efficient thermal management and enhanced light-extraction efficiency (LEE) because of the usage of metal reflectors and highly thermally conductive metal substrates. The LED on Cu, therefore, has stronger photoluminescence, electroluminescence intensities, and better current−voltage characteristics compared to the conventional nanowire LED on Si. Our simulation results further confirm the improved device performance of LEDs on Cu, compared to LEDs on Si. The LEE of the nanowire LED on Cu is nine times higher than that of the LED on Si at the same nanowire radius of 60 nm and spacing of 130 nm. Moreover, by engineering the device-active region, we achieved high-brightness phosphor-free LEDs on Cu with highly stable white-light emission and high color-rendering index of ∼95, showing their promising applications in general lighting, flexible displays, and wearable applications.

INTRODUCTION

III-Nitride semiconductors have been intensively studied for application in optoelectronic devices, because of the superb advantages offered by this material system. The direct energy bandgap III-nitride semiconductors can absorb or emit light efficiently over a broad spectrum, ranging from ~0.65 eV (InN) to 6.4 eV (AlN), which encompasses from a deep ultraviolet to near infrared spectrum. However, owing to the lack of native substrates, conventional III-nitride planar heterostructures generally exhibit very high dislocation densities that severely limit the device performance and reliability. On the other hand, nanowire heterostructures can be grown on lattice mismatched substrates with drastically reduced dislocation densities, owing to the highly effective lateral stress relaxation. The growth of the nearly defect-free III-nitride nanowire heterostructures has been reported on various substrates, including Si and sapphire. Moreover, nanowires have emerged as a powerful platform to effectively scale down the dimensions of the devices and systems, ideally suited for the future nanophotonic and nanoelectronic devices. Nanowire light-emitting diodes (LEDs) with emission in the ultraviolet to visible wavelength range have been intensively studied for the applications in solid-state lighting, flat-panel displays, and solar-blind detectors. However, the currently reported nanowire LEDs generally
exhibit very low external quantum efficiency, which may be attributed to the presence of defects on nanowire surfaces, and/or low light extraction efficiency (LEE). Moreover, III-nitride nanowire LEDs are normally grown on the Si substrates, which may largely absorb the photon emitted from the LED-active region, severely limiting the light output power. Additionally, the Si semiconductor exhibits low electrical conductivity and thermal expansion coefficients compared to a metal substrate. High-power LED applications, however, require large-area chips and can operate at a high injection current which mostly will heat up the devices. Generally, quantum efficiencies, output power, and lifetime reduce rapidly when the junction temperature increases. Therefore, managing heat dissipation is seriously considered. Besides the applications in solid-state lighting illumination, the use of LEDs in telecommunications and decoration displays for the flexible electronics devices has also been extensively developed owing to the feasible integration of such LEDs in these electronic devices.

Organic LEDs have been first studied because of their ease to grow on plastic substrates. However, for these types of LEDs, high electrical performance, low resistance, long-term reliability, and controlled doping concentrations still remain challenging issues. These issues can be addressed in inorganic semiconductors using III–V and related materials. In this regard, the replacement of the Si substrate by a suitable metal, therefore, promises the improved device performance, including high output power and less heating effect. In this paper, we have systematically developed high-brightness InGaN/AlGaN nanowire full-color LEDs on a copper (Cu) substrate from both simulation and experiment. The simulation results show that LEDs on Cu have significantly enhanced LEE compared to the other LED structures, which is ∼9 in 7 times higher than that of LEDs on Si and flip-chip LEDs on the Si substrates, respectively. The experimental results further confirm that the LED on the Cu substrate shows significantly enhanced light output intensity and better current–voltage characteristics over the conventional LEDs on Si substrates. Moreover, highly stable white-light emission with color rendering index of ∼95 was recorded for the flip-chip nanowire LED on the Cu substrate because of the properly controlled indium composition in the device-active region.

**Simulation and Device Structure**

Illustrated in Figure 1, three InGaN/AlGaN nanowire LED structures were comprehensively studied utilizing the finite-difference time domain (FDTD) method. These LED structures include conventional p-side up nanowire LEDs on Si substrates (LED 1), n-side up LEDs on Si substrates (LED 2), and n-side up LEDs on metal substrates (LED 3), shown in Figure 1a–c, respectively. The n-side up LEDs are also called as the flip-chip LEDs throughout this paper.

The device structure is composed of an n-GaN layer, an InGaN/AlGaN multiple quantum well-active region, and a p-GaN layer. The active region is composed of 10 InGaN wells (3 nm-thick layer) sandwiched by 3 nm AlGaN barrier layers. The thicknesses of n- and p-type GaN regions are 350 and 150 nm, respectively. The underlying substrate is assumed to be Si and metal for the typical and flip-chip InGaN/AlGaN nanowire LEDs, respectively. The refractive indices of the AlGaN and n-GaN/p-GaN regions considered in all simulations are 2.6 and 2.5, respectively.

The flip-chip LEDs exhibit lower forward voltage and series resistance, which can be accounted for the enhanced current spreading in the p-GaN layer. This can be attributed to the reason that the whole p-GaN layer is in contact with the Cu layer here, thereby improving the innate high resistivity of the p-GaN, as reported in similar work. Hence, during the design phase of the nanowires, we have further optimized the p-GaN thickness to study the effect of p-GaN thickness on the LEE of nanowire LEDs.

Numerical FDTD solution is employed to numerically investigate and compare the LEE of nanowire LEDs versus the spacing and radius of the nanowires. The measuring monitors placed above and around the LEDs collect all optical power emitted from the designed nanowire device structures. The LEE is defined as the ratio of the power emerging from the structure to the total power generated within the active region by the dipole. Generally, III-nitride nanowire LEDs grown by molecular beam epitaxy (MBE) have radius in the range of 20–80 nm. In our simulation, therefore, we first considered the nanowire array with a hexagonal arrangement of 50 nm radius and the nanowire center-to-center spacing of 130 nm for all LED devices. The geometry of the nanowires has a dominant effect in the emission of generated photons from the active region into air. The radius and spacing of the nanowires are among the parameters which can be used to maximize the LEE of nanowire LEDs. With proper design of nanowire spacing and radius, the light can be coupled through nanowires and scattered out of the device.

Figure 2a,b depict the LEE versus radius at a constant spacing of 130 nm, and LEE versus spacing at a constant nanowire radius of 50 nm, respectively. Shown in Figure 2a, LED 2 demonstrated an enhancement in the LEE compared to LED 1 because of the shorter distance between the active region and the Si substrate, consequently stronger reflection from the substrate. When compared to the light-absorptive properties of the silicon material, the flip-chip LEDs with light-reflective mirrors have been reported to have an enhanced LEE. The aforementioned argument has been used to support the fact that LED 3 has a higher LEE compared to LED 2, attributed to the presence of a stronger reflection of light from the metal/Cu substrate and also because of the reduced absorption in Cu as compared to the Si substrate. The stronger reflection ensures a higher probability of light generated from the active region to be reflected back from the substrate, which is eventually extracted after multiple total internal reflections inside the LED structure. Moreover, the presence of a metal layer in the flip-chip LEDs ensures the formation of a good Ohmic contact at the p-type region and serves as the optical reflector and the current spreading layer to facilitate the current injection to LED-active regions.

![Figure 1](https://example.com/figure1.png)"
Variation of critical parameters such as radius and spacing between nanowires play an important role on LEE. As shown in Figure 2, there is an optimum radius and spacing to avoid photonic bandgap (PBG) and have a high LEE. The electric-field plots depicted in Figure 2c,d show the electric-field distribution for both flip-chip LED structures on Cu and typical LED structures on Si substrate.

Figure 2. Variation of LEE with (a) change in spacing (for a constant radius of 50 nm) and (b) change in radius (constant spacing of 130 nm). (c) Electric-field distribution plot from top monitor for a flip-chip LED on the metal. (d) Electric-field plot from top monitor for a normal p−i−n LED structure on the Si substrate.

Figure 3. (a) LEE for 16 different random structures with different nanowire diameter and nanowire spacing between them. (b) Variation of p-type height with the LEE of a flip-chip (n−i−p) structure on a metal. (c,d) Electric-field contour plots for a typical random flip-chip structure on metal and a random normal (p−i−n) structure on the Si substrate.

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nm and a center-to-center spacing of 130 nm. Illustrated in Figure 2c, in the flip-chip structure, the light is not locally stagnant, rather much better propagated within the structure. In other words, as compared to a conventional LED device composed of multiple nanowires, there is a contribution from the majority of the nanowires within the device toward light extraction, as opposed to a normal p-side up structure on the Si substrate (as shown in Figure 2d), where the light extraction is typically concentrated toward the center and only a minority of nanowires (placed in the middle of the device) contribute to the light extraction.

We have further considered a variation in nanowire radius and wire spacing for nanowire LEDs in which the nanowires are randomly arranged because the self-organized III-nitride nanowires are not naturally periodically arranged on the substrate. Figure 3a represents the LEEs of the 16 typical random flip-chip LED structures with different nanowire radius and spacing. The radius from 47.5 to 52.5 nm and center-to-center spacing in the range of 125–135 nm were considered. The LEE of the random LED structure has an average LEE of 26.79%, whereas that of the nanowire LEDs with nanowire radius of 50 nm and a spacing of 130 nm is 9.61%. For periodic structures, the LED devices may operate in the PBG mode, whereas for the same amount of randomness in a random structure, there is the probability of the device working outside the scope of PBG modes. Figure 3b depicts the effect of the p-GaN thickness on the LEE of a flip-chip device structure. Modification in the p-GaN thickness of the nanowire has a sizable contribution to the LEE. p-GaN shows a strong variation in the trend of LEE with p-type thickness. The latter behavior is explained by the superimposition/interference effect between the forward and backward traveling photons. The latter reaffirms the necessary constraints for constructive reflection occurring within the active region of the nanowire. This in turn stresses the need for optimizing the p-type layer thickness for the best LEE. Compared to the previously mentioned parameters, the p-GaN shows a strong variation in the LEE. It is even worse for smaller thickness because of the high reflection from the reflective layer on the top of the substrate.

Figure 3c,d represents the electric-field contour plots of a random flip-chip LED on the metal and random p-i-n structure on the Si substrate, respectively. As in the case depicted in Figure 2c,d periodic nanowire structures, the distribution of contribution toward light extraction remains the same in the random flip-chip and regular random p-i-n structures, illustrated in Figure 3c.d.

### RESULTS AND DISCUSSION

Highly uniform nanowire LEDs with a radius of ~60 nm were grown on the silicon-on-insulator (SOI) substrate, shown in Figure 4. The device-active region consists of 10 vertically aligned InGaN/AlGaN quantum dots, which can provide white-light emission, because of the In compositional variations (~10–50%) of the InGaN-active region. Each InGaN quantum well has a thickness of ~3 nm and is capped by ~3 nm AlGaN layer. Detailed growth conditions for such nanowire LED heterostructures were described in previous publications.

The surface morphology and orientation of the nanowire LEDs grown directly on the SOI substrate are much more uniform compared to those of the nanowire LEDs grown on the SiO2-on-Si substrate reported in our previous study because of the local surface roughness of amorphous SiO2-on-Si. Such properties may lead to higher optical and structural performances of the LEDs on SOI compared with LEDs on the SiO2-on-Si wafers.

As shown in Figure 5a, after being transferred to the Cu substrate, the photoluminescence (PL) intensity of the nanowire LED on Cu (LED 3) is enhanced by a factor of 1.5 compared to that of the as-grown nanowire nanowires on the SOI substrate. Such an enhancement is mainly attributed to the improved light extraction efficiencies because of the significantly reduced light absorption after removing the Si substrate. Moreover, the Ni/Au bilayer may work efficiently as a reflector to further enhance the LEE. This further confirms that the nanowire damage during the transferring process is almost negligible. A small blue shift of ~1.5 nm was recorded for the nanowire LED on Cu compared to that of the nanowire LED on the SOI substrate, which is attributed to the fact that the strain has been relaxed owing to the release of nanowires from the SOI substrate. The electroluminescence (EL) intensity of LED 3 was measured nearly twice higher than that of the regular nanowire LED on Si (LED 1), as shown in Figure 5b. Such enhanced PL and EL intensities agree well with the simulation results presented in Figures 2 and 3.

Figure 6a shows the current–voltage characteristics of a nanowire LED on Cu compared to a similar growth condition nanowire LED on Si substrates. The LEDs on the Cu substrate (LED 3) exhibit excellent current–voltage characteristics and show lower leakage current and slightly higher current in forward bias, compared to the LED device on the Si substrate (LED 1) at the same voltage. At 20 mA injection current, the operating voltages for LED 3 and LED 1 are 3.2 and 4.1 V, respectively. At ~8 V reversed bias, the corresponding currents for LED 3 and LED 1 are −0.5 and −1.5 mA, respectively. The lower voltage of LED 3 may be attributed to the better heat dissipation and substrate conductivity of the nanowire LED on the Cu substrate, which is consistent with other reports. As shown in the inset of Figure 6a, the optical image of light emission from the LED device on the Cu substrate demonstrates the successful fabrication of such a nanowire on metal substrates. Figure 6b shows the relative light output power of both LED 1 and LED 3 for comparison. It is shown that LED 3 exhibits stronger output power compared to that of LED 1. Such an output power enhancement is attributed to the enhanced LEE in LED 3, which was explained previously. To further increase the output power of LED 3, several limiting factors should be considered and addressed, for instance, improving the transparency of the top Ti/Au/ITO metal contact and optimizing the nanowire geometry including wire density, diameter, and height.
We have further developed phosphor-free white LEDs on the Cu substrate by engineering the In composition in the InGaN-active region, thereby, white-light emission with full visible spectrum was achieved. This method was reported elsewhere.\textsuperscript{16} Figure 7a shows the EL spectra of the phosphor-free white LEDs on the Cu substrate under different injection currents. The experiment was conducted under pulse bias condition with a duty cycle of 1\% to minimize the junction heating effect. The peak wavelength is almost stable when the current increases from 50 to 500 mA, attributed to the low-quantum-confined Stark effect.\textsuperscript{50} Moreover, the LED device achieves stable white-light emission on the CIE diagram, shown in Figure 7b. The device achieves a high CRI of \sim95 showing a promise as a candidate for solid-state lighting and display.

\section*{CONCLUSIONS}

In summary, we have demonstrated the high-performance InGaN/AlGaN nanowire white LEDs on the Cu substrate without using any phosphor converter. The improved performance including stronger PL and EL intensities, and current-voltage behavior were recorded for the transferred nanowire LED on the Cu substrate, compared to the conventional LED on Si. Such enhancements are attributed to the reduced light absorption by the substrate, the enhanced LEE, and the reduced heating effect. Moreover, the substrate-transfer approach presented in this paper can be applicable for fabricating the LED devices on metal substrates, plastics, or many other platforms where the epitaxial growth of the LED structures cannot be directly grown on, because of their high growth temperature. This work also provides a promising...
approach for the integration of III-nitride nanowire LEDs in future solid-state lighting, visible light communication, and decoration displays as flexible electronic devices.

■ EXPERIMENTAL SECTION

MBE Growth. The self-organized InGaN/AlGaN nanowire LED heterostructures were grown on the Si and SOI substrates by radio frequency plasma-assisted Veeco Gen II MBE. The SOI wafer structure includes 50 nm Si on 2 μm SiO2 and 725 μm Si in which the SiO2 serves as an etch-stop layer during the Si-etching process. The nitrogen flow was kept at 1.0 standard cubic centimeter per minute (scm) during the epitaxial growth process and a forward plasma power of ~350 W. The n-GaN and p-GaN were grown at ~770 °C, whereas the InGaN/AlGaN-active region was grown at 640–680 °C to enhance the In incorporation. The device active region consists of 10 vertically aligned InGaN/AlGaN quantum dots, which can provide white light emission because of the In compositional variations (~10–50%) of the InGaN-active region.

Device Fabrication. The fabrication process of LEDs on the Cu substrate is described in Scheme 1. The nanowire LEDs were first grown on the SOI substrate by MBE. A polyimide resist was then spin-coated to fully cover the nanowire, followed by oxygen plasma etching to reveal the nanowire top portions (Scheme 1a). Subsequently, polyimide was hard-baked at 350 °C for 45 min. Metal contact with Ni (10 nm)/Au (150 nm) was then deposited on top of the nanowires to serve as the p-type contact and also the metal reflector, followed by electroplating of 150 μm Cu. Deep reactive-ion etching was then applied to remove the Si substrate with the etching rate of 12 μm/min, illustrated in Scheme 1b. The etch-stop SiO2 layer was then removed by buffered oxide etching solution, whereas the remaining Si top layer was removed by tetra methyl ammonium hydroxide. Finally, Ti (5 nm)/Au (5 nm)/ITO (200 nm) and Ti (10 nm)/Au (100 nm) were deposited to serve as the top-metal contacts, shown in Scheme 1c. The LEDs on the Si substrate were also fabricated for comparison. Devices with areal size of 1 × 1 mm² were used for characterization. For a fair comparison, all measurements were performed on the LEDs at similar locations on the wafers for both LEDs on Cu and LEDs on Si substrates.

Characterization. Detailed optical and electrical characterizations of the nanowire LEDs on the Cu substrate were performed. PL study was performed by using a 405 nm laser as the excitation source with a microscope objective, a high-resolution spectrometer, and a photomultiplier tube to collect and detect the emissions from the LED samples. The current–voltage measurements of the nanowire LEDs were measured using a power supply (Keithley 2402). EL emission of the LED devices was collected by an optical fiber directly connected to an Ocean Optics spectrometer. All measurements were conducted at room temperature.

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The manuscript was written through contributions of all authors.

Notes
The authors declare no competing financial interest.

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