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Bryan Melanson, Cheng Liu, Matthew Hartensveld, Jing Zhang, "Light extraction analysis of AlGaN nanowires with inverse taper for DUV LEDs," Proc. SPIE 11706, Light-Emitting Devices, Materials, and Applications XXV, 117060I (5 March 2021); doi: 10.1117/12.2583116

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Event: SPIE OPTO, 2021, Online Only

Light Extraction Analysis of AlGaN Nanowires with Inverse Taper for DUV LEDs

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ABSTRACT

Planar deep-ultraviolet (DUV) light emitting diodes (LEDs) suffer from extremely low external quantum efficiencies (EQEs) due to poor light extraction efficiencies (LEE) which are often less than 1%, hindering their widespread use. In AlGaN DUV LEDs with high Al-content, the positioning of the valence subbands leads to dominant transverse magnetic (TM)-polarized emission which is difficult to extract from planar devices. To improve the LEE of DUV LEDs, techniques such as surface roughening and nanowire formation have been used. Nanowires are especially promising for DUV LEDs because they allow for very efficient extraction of TM-polarized light through their sidewalls. In this work, we demonstrate a novel "inverse taper" profile in AlGaN nanowires, in which the base of the nanowire can be narrowed to have a smaller diameter than the top through a KOH-based wet etch process. Hydroxyl-based chemistries are known to have a lower etch rate against the c-plane of wurtzite AlGaN alloys. Here, we report on observations of 0.8% KOH at 80°C exhibiting a unique selectivity to a different wurtzite crystal plane, believed to be the $(20\bar{2}1)$ plane, allowing for formation of an inverse taper structure. Finite difference time domain (FDTD) simulations at 280 nm reveal that AlGaN nanowire LEDs with high sidewall inverse taper angles can have greater than 75% and 90% LEE for TE and TM-polarized light respectively, ~2.5x higher than the LEE of vertical sidewall nanowires. This novel phenomenon may allow for significant improvements in the LEE of DUV nanowire LEDs.

Keywords: AlGaN, Nanowire, Ultraviolet, LED, Efficiency, Taper, Light Extraction Efficiency, Solid State Lighting

1. INTRODUCTION

Deep Ultraviolet (DUV) LEDs show great promise for use in applications such as water purification, surface sterilization, medical treatments, resin curing, and high density optical data storage[1–4]. In comparison to the mercury vapor arc lamps which they stand to replace, DUV LEDs offer tunable emission wavelength, longer lifetime, greater stability, smaller form factors, environmental compatibility, increased physical durability, and much lower power consumption and heat generation[1,5]. DUV LEDs are typically fabricated as mesa structures, with dimensions ranging from tens to hundreds of microns. While this fabrication approach works well for visible light LEDs emitting between 400 and 700 nm, mesa structures severely restrict the light extraction efficiency (LEE) of DUV emitting epi-stacks due to differences in the polarization of the DUV light emitted from the active region. While visible light LEDs based on GaN and InGaN epi-stacks emit almost 100% of their light with transverse electric (TE) polarization (electric field component parallel to the quantum well (QW) layer), AlGaN-based DUV LEDs emitting below 300 nm also produce a non-negligible fraction of their light with transverse magnetic (TM) polarization (magnetic field component parallel to the QW layer). At wavelengths below ~240 nm, TM-polarized emission even becomes dominant due to complex valence band mixing effects which occur as the Al-content of the AlGaN QW layers is increased[6]. TM-polarized light is exceptionally difficult to extract from mesa structures due to its increased propensity to propagate in the plane of the epi-stack where it is readily reabsorbed, rather than escaping out of the mesa structure through the top as is the case with TE-polarized light. The LEE of mesa LEDs emitting non-negligible fractions of TM-polarized light is consequently extremely poor, often less than 1%, compared to values of more than 70% for blue LEDs[7,8].

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Light-Emitting Devices, Materials, and Applications XXV, edited by Jong Kyu Kim, Michael R. Krames, Martin Strassburg, Proc. of SPIE Vol. 11706, 117060I · © 2021 SPIE
CCC code: 0277-786X/21/$21 · doi: 10.1117/12.2583116

To improve the LEE of DUV LEDs, advanced fabrication techniques such as surface roughening[9], substrate patterning[10], diffraction gratings[11], and photonic crystals[12] have been implemented with reasonable success. Another approach which has seen very little investigation is the use of nanowire arrays LEDs to improve LEE. By fabricating arrays of narrow, vertical nanowires with the DUV emitting QWs located near the tips of the wires, it is possible to massively improve the extraction of TM-polarized light, as the in-plane propagation distance is significantly reduced, eliminating reabsorption. We have previously demonstrated that hydroxyl-based chemistries are capable of selectively etching GaN and InGaN, removing dry etch damage and "straightening" the sidewalls of the vertical GaN structures[13]. This is possible because the OH^- ions in solution preferentially etch the non-polar and semi-polar planes of wurtzite GaN, while leaving the Ga-polar c-plane ((0001)) untouched[14]. GaN and its alloys are typically etched using Cl-plasma, with the resulting dry etched structures usually exhibiting a slight "normal taper". Hydroxyl based etches are capable of removing this taper, as the etch chemistry will rapidly etch the exposed semi-polar $(10\bar{1}1)$ plane until the non-polar $(10\bar{1}0)$ plane is revealed, allowing for formation of structures with perfectly vertical sidewalls[13]. This behavior has been extensively documented in GaN to the extent that it has been adopted in commercial fabrication of GaN LEDs and power electronics. By comparison, very little research exists on the wet etching of AlGaN, especially the high Al-content AlGaN used in DUV LEDs, with hydroxyl-based chemistries[15].

Here we report on a unique phenomenon in which high Al-content AlGaN ($Al_{0.55}Ga_{0.45}N$) nanowires are shown to develop an "inverse taper" profile (base diameter smaller than top diameter) when subjected to extended etches in ~0.8 wt% KOH at 80°C. While the exact mechanism behind the phenomenon is unknown, we believe that a unique selectively to a different crystal plane, possibly the $(20\bar{2}1)$ or $(10\bar{1}1)$ plane, may exist for high Al-content AlGaN and be responsible for this as-of-yet unreported behavior. We report on the time dependence of the inverse taper angle (α), showing it to be roughly linear, reaching a value of 11° after 70 min in the aforementioned heated etch solution. A finite difference time domain (FDTD) analysis of the effects of this inverse taper angle on the LEE of the nanowires is also performed.

2. EXPERIMENTAL RESULTS

2.1 Nanowire Fabrication

A commercially grown 50 mm AlGaN epi-wafer was used to fabricate the nanowires. The epitaxial stack was grown on a 430 µm thick sapphire substrate using metal organic chemical vapor deposition (MOCVD), and consisted of a 1 µm AlN buffer layer, 20x AlN/AlGaN super lattice, and various AlGaN layers. The complete epi-stack is shown in Fig. 1. After a standard RCA clean, LOR 5A liftoff resist and AZ MiR 701 photoresist were coated onto the wafer and exposed using a Heidelberg DWL-66+ laser direct write (LDW) system.

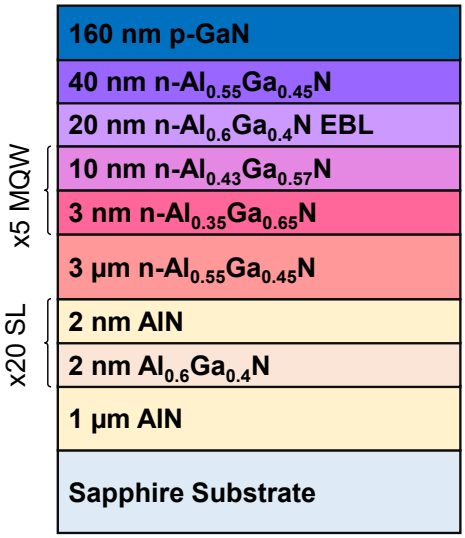

Figure 1. Cross sectional schematic of the wafer epitaxial stack used for fabrication of the AlGaN nanowires. Layer thickness not to scale.

Ni was then deposited using electron beam physical vapor deposition to a thickness of 200 nm and lifted of in N-Methyl-2-Pyrrolidone (NMP), leaving arrays of 2 µm Ni dots on the surface of the wafer (Fig. 2(b)). These Ni dots acted as a hard mask for the Cl plasma dry etch which was used to form the nanowires. A Plasma-Therm Apex inductively coupled plasma (ICP) etcher was used to perform the dry etch (Fig. 2(c)). Gas flow rates of 40 standard cubic centimeters per second (sccm) of Cl_2 and 10 sccm Ar at an etch chamber pressure of 30 mtorr were used, along with and ICP plasma power of 500 W and a RIE forward power of 225 W. The resulting nanowires were approximately 2.5 µm tall and 2 µm in diameter, and exhibited a cone-like "normal taper" with their bottom diameter larger than the 2 µm top diameter.

Following the dry etch, the nanowires were subjected to the hydroxyl-based wet etch. AZ400K, a positive photoresist developer containing 2 wt% KOH was used as the source of OH^- ions. An aqueous 40% AZ400K solution (0.8 wt% KOH) was prepared and heated to a stable 80°C on a hotplate. The wafer was immersed in the solution for 5 min and then removed and rinsed thoroughly in deionized (DI) water for 3 minutes to remove etch byproducts from the wafer surface. The nanowires were then imaged using a scanning electronic microscope (SEM). This process of etching and imaging was repeated a total of five times in order to record the inverse taper angle α after 5, 10, 20, 30, 50, and 70 min (Figs. 2(d-e)). Fig. 2 shows a general schematic of the fabrication process.

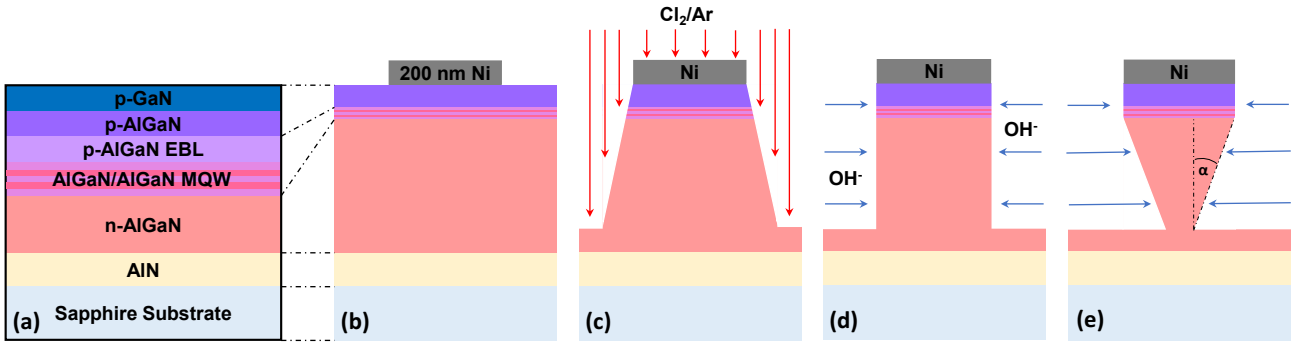

Figure 2. Cross section schematic of the nanowire fabrication and wet etching process. (a) A simplified version of the AlGaN epitaxial stack shown in Fig. 1, (b) patterning of the Ni hard mask onto the wafer, (c) dry etching of the nanowires using a Cl_2/Ar plasma, (d) wet etching of the nanowires using a heated 0.8 wt% KOH solution, (e) resulting nanowire "inverse taper" morphology following extended KOH etching.

2.2 Nanowire Characterization

SEM images were taken of the nanowires at off normal angles in order to determine the inverse taper angle, as shown in Fig. 3. In contrast to GaN nanowires and other nanostructures, AlGaN nanowires show a distinct undercut which increases with etch time. The sidewalls of both GaN and AlGaN nanostructures have a sidewall taper angle $\alpha < 0°$ following the dry etch, and in the GaN/InGaN materials system α approaches 0° as the wires are etched in hydroxyl-based chemistry.

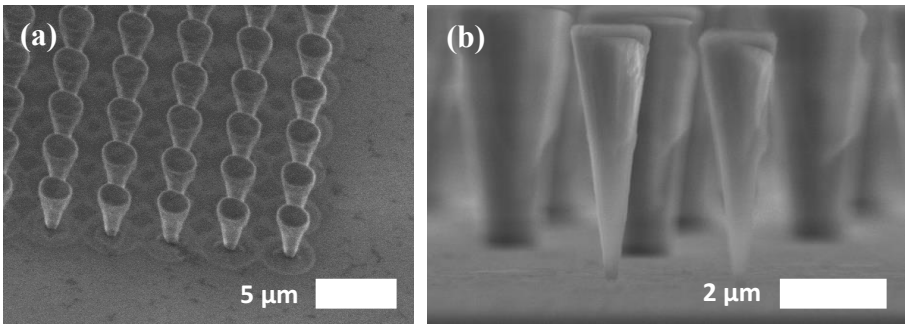

Figure 3. (a) Representative SEM image (45°) of nanowires after 50 min in the KOH etchant. (b) SEM image (87°) showing nanowires with smaller top diameters nearly separated from the substrate after 70 min in the KOH etchant.

Unlike GaN/InGaN nanostructures, whose taper angle α never exceeds 0° even for etch times in excess of 1 hour, AlGaN nanowires develop a taper angle of α > 0° after only a few minutes in the etch solution, as can be seen in Fig. 3(a). This indicates that the mechanism for hydroxyl-based wet etching of AlGaN is slightly different than for GaN/InGaN. As with GaN/InGaN the c-plane (top) of the nanowires is unaffected by the wet etch, and the wires maintain the same height of 2.5 µm for the duration of the wet etch, up to 70 min. Fig. 4 shows the taper angle α as a function of etch time. The relationship between α and etch time is roughly logarithmic, and α appears to approach ~12-16°. Some of the nanowires on the wafer had reduced diameters, down to ~1 µm, due to lithography issues. The taper angle was found to be independent of the wire diameter, and as a consequence, wires which started with small enough top diameters due to lithography errors were removed from the substrate, as a taper angle of α = 11°, achieved after 70 min in etch solution, was sufficient to narrow the base of these wires to a point, causing them to separate from the substrate. Fig. 3(b) shows nanowires with top diameters of ~1.5 µm etched for 70 min until they are nearly removed from the substrate.

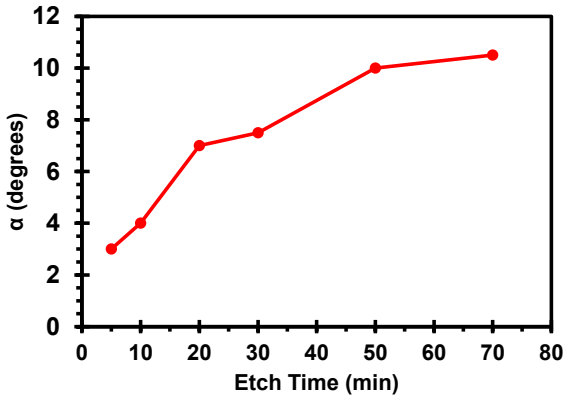

Figure 4. AlGaN nanowire taper α angle as a function of etch time.

The inverse taper profile is caused by the crystallographic selectivity of the hydroxyl-based etch. Ga polar surfaces effectively repel the OH^- ions with N-polar, semi-polar, and non-polar planes are etched more readily[13]. In GaN nanostructures, a 1:1 Ga:N ratio exists, and is responsible for the well documented wet etch behavior described previously. The etch proceeds according to a cavity model, in which GaN is oxidized to Ga_2O_3 by the OH^- ions in solutions, with the water soluble Ga_2O_3 then being removed in the aqueous etch solution. In AlGaN, Al atoms occupy some of the Ga sites in the crystal lattice. In the case of our $Al_{0.55}Ga_{0.45}N$ nanowires, 55% of the Ga sites are occupied by Al atoms. The Al:N covalent bond is stronger than the Ga:N covalent bond (11.5 eV/atom vs 8.9 eV/atom), and as such a larger energy barrier exists for oxidation of AlGaN[16]. This likely alters the etch rate against certainly crystallographic planes, as each plane has a different areal density of Al, Ga, and N atoms and consequently a different polarity and resistivity to oxidation of OH^- ions in solution. In our AlGaN nanowires, it appears the taper angle approaches ~15°, which is very close to the off-normal angle of the wurtzite $(20\bar{2}1)$ plane (16°). As such, it is likely that the $(20\bar{2}1)$ may limit the progression of the etch in AlGaN just as the $(10\bar{1}0)$ plane does in GaN/InGaN. Further experimentation is needed to confirm this and deduce the exact mechanisms behind this unique phenomenon observed only in AlGaN.

3. LIGHT EXTRACTION ANALYSIS

3.1 Simulation Setup

Three-dimension FDTD simulations were performed in order to investigate the effects of a positive taper angle (α > 0°) and nanowire height on the LEE of AlGaN nanowires using Synopsys Fullwave[17]. TE and TM polarized emission were investigated separately. A cross sectional schematic of the 3D structure used for the simulations is shown in Fig. 5. Perfectly matched layer (PML) domain boundary conditions were used on all domain surfaces in order to capture the most accurate results for LEE[18]. The height, top diameter, and taper angle of the nanowire were defined manually, with the bottom diameter being dependent upon these three parameters. Nanowires with a diameter of 1 µm, taper angles between -10° and 25° degrees, and heights between 100 nm and 3.5 µm were investigated extensively at an emission wavelength

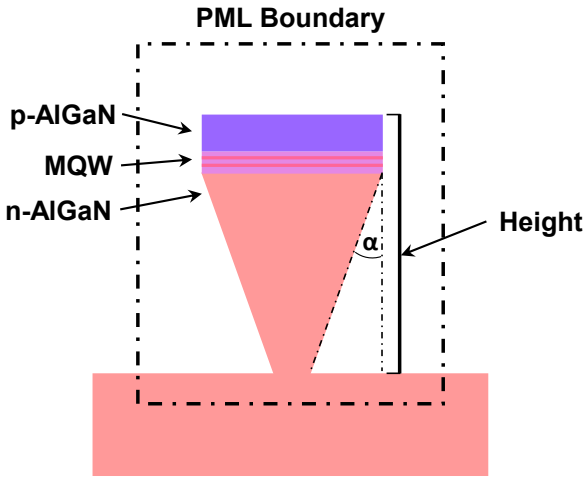

Figure 5. Cross-sectional schematic of the structure used for FDTD simulation of AlGaN nanowires. Shown cross section is radially symmetric in 3D.

of 280 nm. A single emission dipole source was used for all simulations and was located in the center of the nanowire in the middle of the MQW active region.

3.2 Simulation Results

Fig. 6(a) plots the LEE of nanowires of varying height as a function of the taper angle α for both TE and TM polarized emission at 280 nm. As all simulated wires have a diameter of 1 μm, it follows that shorter wires are capable of exhibiting higher taper angles, as is evident from Fig. 6(a). It is evident that the LEE increases monotonically for wires of all heights, for both TE and TM polarizations, indicating that an inverse taper profile will enhance the light extraction from AlGaN nanowires. It is also evident that taller wires can achieve much higher LEE at smaller α, for example, the TM LEE of 2.5 μm tall nanowires at 10° is ~92%, compared to only 66% for 1 μm wires at 10°. This is likely because taller wires offer light more opportunities to escape before being reflected into the substrate where it is reabsorbed. This result is extremely important because it indicates the LEE can be significantly enhanced my simply making the nanowires taller. Likewise, wires with higher α have narrower base diameters, reducing the cross section through which light can travel into the substrate. Angled sidewalls also aid in directing light out the top of the nanowire.

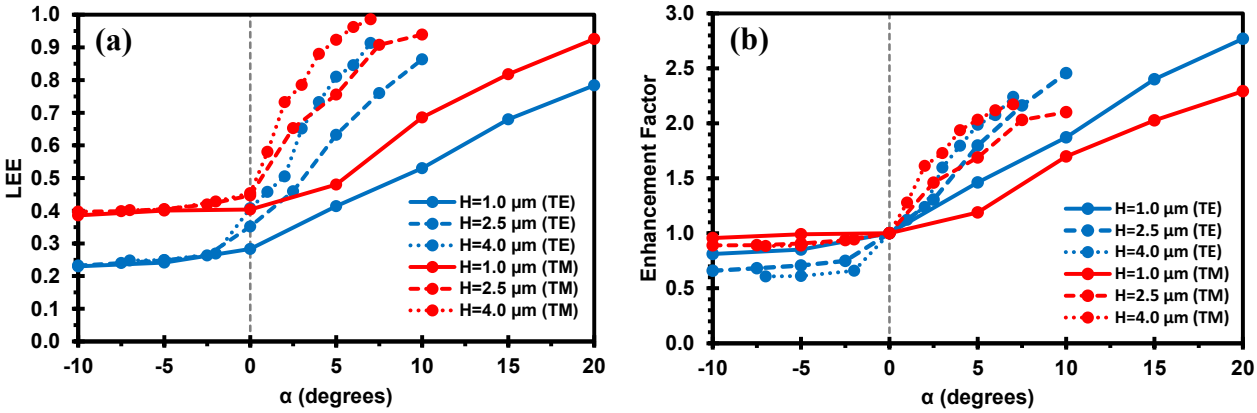

Figure 6. LEE (a) and LEE enhancement factor (b) as a function of inverse taper angle α for both TE (blue) and TM (red) polarizations and wire heights of 1.0 μm (solid), 2.5 μm (dashed), and 4.0 μm (dotted), at an emission wavelength of 280 nm. All wires have a top diameter of 1 μm.

Fig. 6(a) also indicates that TM-LEE is generally higher than TE-LEE. This is due to the propensity of TM polarized photons to propagate within the epitaxial plane of the QW and surrounding epi-layers. In a wide, mesa LED structure, this TM light usually propagates until it is reabsorbed, however, in nanowires, especially those with $\alpha > 0°$, this light can easily exit through the sides of the nanowire. Fig. 6(b) shows the data from Fig. 6(a) replotted in terms of the LEE enhancement factor, which is derived by dividing the LEE of a wire at any point by the LEE of that wire at $\alpha = 0°$. From this it is evident that enhancements in excess of 2x can be achieved for all wire heights for both TE and TM polarization. Taper angles of $\alpha < 0°$ (normal taper) serve to decrease the LEE and LEE enhancement factor, as these "cone-like" structure serve to increase the base cross section of the wire, allowing more light to travel into the substrate. This effect can be visualized in Fig. 7, which shows cross sectional spatial electric field intensity plots of nanowires 1 µm in diameter with heights of 1 µm and 2.5 µm at various values of α. Nanowires with $\alpha > 0°$ are seen to exhibit higher electric field intensity in the free space surrounding them than those with $\alpha < 0°$ and $\alpha = 0°$, indicating the more light has escaped from the wires. The narrower base diameters of these wires serve to reduce the cross-sectional area through which light can escape into the substrate, as can be clearly seen in Fig. 7. Nanowires with larger α also develop more complex internal optical modes, which aids in directing light out into free space.

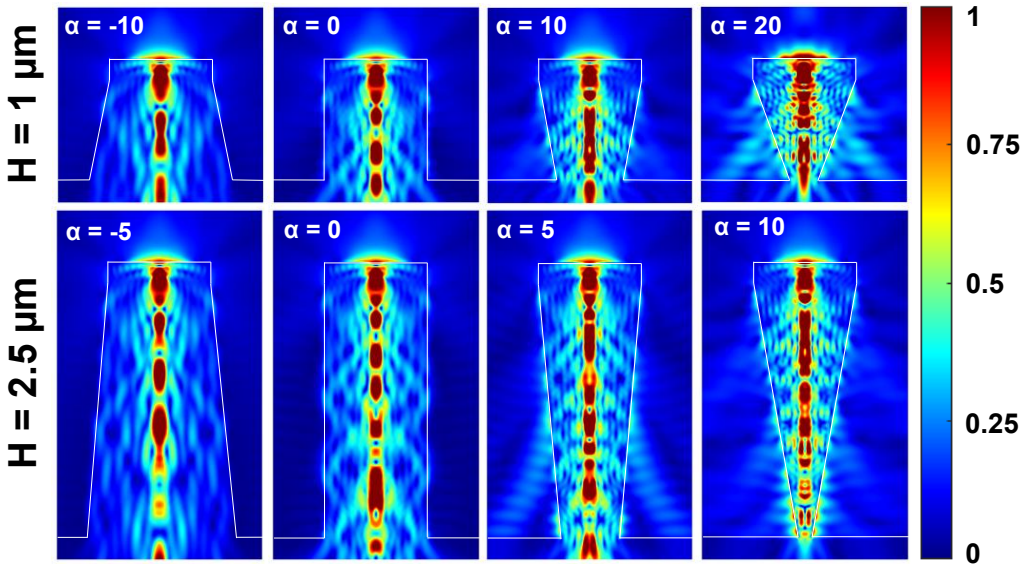

Figure 7. Cross sectional electric field intensity plots of 1 µm and 2.5 µm tall nanowires with taper angles between -10° and 20° and -5° and 10°, respectively.

Fig. 8 shows both the TE (Fig. 8(a)) and TM (Fig. 8(b)) LEE as a function of nanowire height at $\alpha = 10°$. As indicated by Fig. 6, LEE increases monotonically with nanowire height for both TE and TM polarized emission. Narrower nanowires generally have higher LEE, as it is easier for light to escape when it doesn't have to travel as far to reach the surface of the nanowire. The LEE of wires of all three diameters approaches or exceeds 90% at their maximum height, with the general trend being that taller wires are more efficient at extracting light. The data presented in Figs. 6-8 show that an inverse taper profile can be extremely effective at enhancing the LEE of AlGaN nanowires emitting at 280 nm.

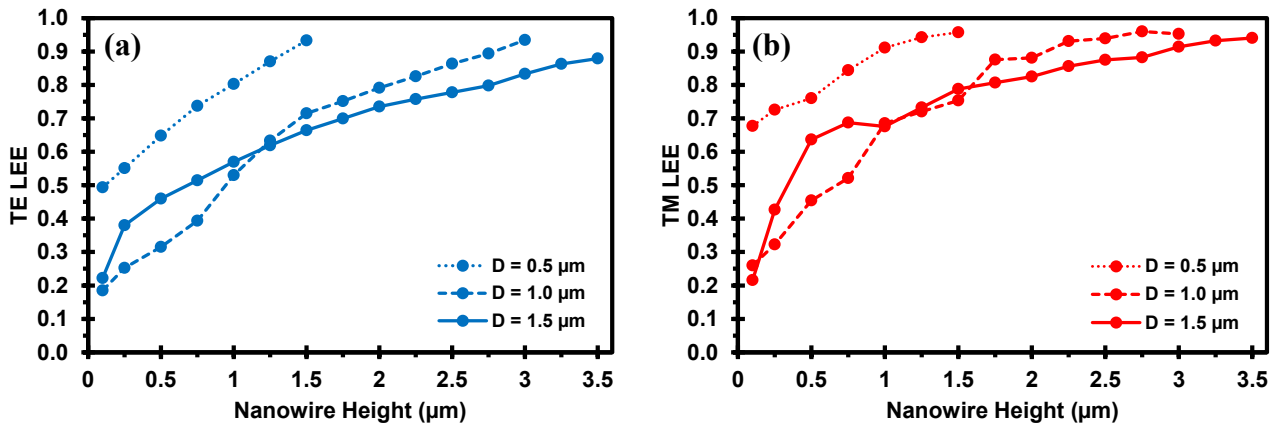

Figure 8. TE (a) and TM (b) LEE as a function of nanowire height for nanowires with diameters of 0.5 μm (dotted), 1.0 μm (dashed), and 1.5 μm (solid).

4. CONCLUSIONS

This work documents for the first time a unique phenomenon in the AlGaN materials system in which extended wet etches in 0.8 wt% KOH at 80°C produce a distinct "inverse taper" profile in vertical dry etch structures. This inverse taper was demonstrated on 2 μm diameter $Al_{0.55}Ga_{0.45}N$ nanowires ~2.5 μm tall, with the inverse taper angle α increasing with etch time up to a value of ~11° after 70 min. From these preliminary results we estimate that the taper angle is asymptotic to 16°, which corresponds to the $(20\bar{2}1)$ plane in wurtzite AlGaN. This unique crystallographic etch selectivity is likely due to the higher binding energy of the Al:N bond and altered polarity of certain crystal planes, causing the KOH etch to behave differently than it does against GaN and InGaN. 3D FDTD simulations were performed in order to investigate the effects of this unique inverse taper structure on the LEE of AlGaN nanowires. Results show that both TE and TM LEE increase monotonically with increasing taper angle, as well as with increasing nanowire height, with LEE enhancements of up to 2.5x achievable for the maximum allowed undercut angle for most nanowire geometries when compared to the case of α = 0°. Combined, the experimental and simulation results presented here indicate that this unique phenomenon may allow for significant enhancements in the LEE and overall external quantum efficiency of DUV AlGaN nanowire LEDs, making them more viable in a variety of applications.

ACKNOWLEDGMENTS

The authors would like to acknowledge funding support provided by the National Science Foundation under Award No. ECCS 1751675.

REFERENCES

[1] Kneissel, M., and Rass, J., [III-Nitride Ultraviolet Emitters: Technology and Applications] , Springer International Publishing (2016).
[2] Hirayama, H., Maeda, N., Fujikawa, S., Toyoda, S., and Kamada, N., "Recent progress and future prospects of AlGaN-based high-efficiency deep-ultraviolet light-emitting diodes" 53(10), 100209 (2014).
[3] Liu, C., Melanson, B., and Zhang, J., "AlGaN-Delta-GaN Quantum Well for DUV LEDs," Photonics 7(4), (2020).
[4] Jarvis, P., Autin, O., Goslan, E., and Hassard, F., "Application of Ultraviolet Light-Emitting Diodes (UV-LED) to Full-Scale Drinking-Water Disinfection," Water 11(9), 1984 (2019).

[5] Nakamura, S., and Krames, M., "History of Gallium–Nitride-Based Light-Emitting Diodes for Illumination," Proceedings of the SPIE 101(10), 2211 (2013).
[6] Rodrigues, S.C.P., Scolfaro, L.M.R., Leite, J.R., and Sipahi, G.M., "Valence band structure of cubic AlGaN/GaN superlattices," Applied Physics Letters 76(8), 1015–1017 (2000).
[7] Manikandan, M., Nirmal, D., Ajayan, J., Mohankumar, P., Prajoon, P., and Arivazhagan, L., "A review of blue light emitting diodes for future solid state lighting and visible light communication applications," Superlattices and Microstructures 136, 106294 (2019).
[8] Shakya, J., Kim, K.H., Lin, J.Y., and Jiang, H.X., "Enhanced light extraction in III-nitride ultraviolet photonic crystal light-emitting diodes," Applied Physics Letters 85(1), 142–144 (2004).
[9] Hirayama, H., Maeda, N., and Lee, J.I., "FDTD analysis of the light extraction efficiency of OLEDs with a random scattering layer," Optics Express 22(1), 498–507 (2014).
[10] Ooi, Y.K., and Zhang, J., "Light Extraction Efficiency Analysis of Flip-Chip Ultraviolet Light-Emitting Diodes With Patterned Sapphire Substrate," IEEE Photonics Journal 10(4), (2017).
[11] Lim, T.B., Cho, K.H., Kim, Y.H., and Jeong, Y.C., "Enhanced light extraction efficiency of OLEDs with quasiperiodic diffraction grating layer," Optics Express 24(16), 17950–17959 (2016).
[12] Lee, Y.J., Kim, S.H., and Do, Y.R., "A high-extraction-efficiency nanopatterned organic light-emitting diode," Applied Physics Letters 82, 3779–3781 (2003).
[13] Hartensveld, M., Ouin, G., Liu, C., and Zhang, J., "Effect of KOH passivation for top-down fabricated InGaN nanowire light emitting diodes," Journal of Applied Physics 126(18), 183102 (2019).
[14] Yu, F., Yao, S., Romer, F., and Wasisto, H.S., "GaN nanowire arrays with nonpolar sidewalls for vertically integrated field-effect transistors," Nanotechnology 28(9), 095206 (2017).
[15] Chen, W., Lin, J., Hu, G., and Zhang, B., "GaN nanowire fabricated by selective wet-etching of GaN micro truncated-pyramid," Journal of Crystal Growth 426, 168 (2015).
[16] DeVre, M., and Westerman, R., "Advances in GaN Dry Etching Process Capabilities"5.
[17] [Fullwave] , Synopsys Optical Solutions Group (2021).
[18] Berenger, J.P., "A perfectly matched layer for the absorption of electromagnetic waves," Journal of Computational Physics 114(2), 185–200 (1994).