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150 mW deep-ultraviolet light-emitting diodes with large-area AlN nanophotonic light-extraction structure emitting at 265 nm

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High-power 265 nm deep-ultraviolet (DUV) AlGaN-based light-emitting diodes (LEDs) with large-area AlN nanophotonic light-extraction structures that were fabricated by a nanoimprint lithography process are presented. Each DUV-LED has a large active area (mesa size of $\sim$0.35 mm^2) and a uniform current spreading design that allows high injection current operation. We have shown that these DUV-LEDs with their large-area nanoimprinted AlN nanophotonic structures exhibit wider near-field emitting areas, stronger far-field extracted light intensities, and an approximately 20-fold increase in output power when compared with a conventional flat-surface DUV-LED. A large-area nanoimprinted single-chip DUV-LED operating in the UV-C wavelength regime has demonstrated a record continuous-wave output power in excess of 150 mW for an injection current of 850 mA at a peak emission wavelength of 265 nm. © *2017 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/).* [http://dx.doi.org/10.1063/1.4978855]

For rapid penetration of the potential applications and markets for aluminum gallium nitride (AlGaN)-based deep-ultraviolet (DUV) light-emitting diodes (LEDs), which include air/water purification, surface disinfection, chemical and biological agent detection, lithographic microfabrication, and medical diagnostics,[1–3] it will be necessary to achieve higher LED output powers and to minimize the total DUV-LED fabrication cost. However, despite numerous development efforts,[4–7] DUV-LEDs continue to have much lower output powers from single chips than the more widely used blue LEDs.[8] The main causes of the low output powers and low external quantum efficiencies (EQEs) of DUV-LEDs are high dislocation densities in the AlGaN-based active layers[1] and the extremely low light extraction efficiency of these devices. AlGaN-based DUV-LEDs are typically grown on sapphire substrates.[4–6] The large lattice and thermal expansion coefficient mismatches between these substrates and the epitaxial layers causes large numbers of dislocations and poor device reliability. To overcome the dislocation problem, we recently reported DUV-LEDs with low dislocation densities of less than 10^6 cm^{-2} that were fabricated on AlN substrates for operation in the DUV region.[9,10] Therefore, the most critical remaining barrier to widespread use of DUV-LEDs is the cost-effective light extraction technology to enhance the output powers of these devices.

Flip-chip designs are generally used for AlGaN-based DUV-LEDs because conventional LED contact layer materials are highly absorbing in the DUV regime. The DUV light emitted by the active layer is thus extracted through a transparent substrate such as sapphire to minimize light absorption by the *n* and *p* contact layers that are located on the epitaxial surface. The low light extraction and low output power of flip-chip DUV-LEDs on AlN substrates is mainly caused by total internal reflection at the interface between the AlN surface and the ambient medium and by internal optical absorption by the *p*-GaN contact layer and the bulk AlN substrate. While GaN strongly absorbs DUV light, a *p*-GaN contact layer is required to provide a good ohmic contact and to reduce the series resistance for reliable high-power LED operation. Additionally, while the AlN substrate solves the dislocation problem, the high refractive index of AlN ($n_{\mathrm{AlN}} \sim 2.3$) leads to a much smaller critical angle (i.e., a very narrow light escape cone) than that of conventional sapphire substrates ($n_{\mathrm{sapp}} \sim 1.8$), which is a serious drawback for light extraction. The bulk AlN substrate also generally has intrinsic optical absorption in the DUV region caused by impurities. AlN substrate fabrication by hydride vapor-phase epitaxy (HVPE) can significantly reduce the optical absorption when compared with that of typical AlN substrates fabricated by physical vapor transport (PVT) with an absorption coefficient of 35 cm^{-1},[7] but HVPE-AlN substrates still show inescapable absorption losses, with an absorption coefficient of approximately 10 cm^{-1} at a wavelength of approximately 265 nm.[10] Therefore, a trade-off exists between the dislocation density and light extraction. Additionally, early power saturation (i.e., efficiency droop) at high injection current densities is a common problem in flip-chip mesa-type DUV-LEDs.[11,12] Therefore, to achieve both high reliability and high power operation, it is essential to enlarge the light escape cones of flip-chip DUV-LEDs on AlN substrates to minimize optical absorption while simultaneously expanding the emitting area to allow higher current operation and increased output flux per single chip LED. In this letter, we report on the design and large-area fabrication of sub-270 nm AlGaN-based flip-chip DUV-LEDs on AlN substrates with light-extraction structures to improve the LED output power.

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A light-extraction structure design that used an AlN nanophotonic structure was previously demonstrated for conventional small-area LEDs.[13] These structures were fabricated by electron-beam (EB) lithography, which is a high-precision but seriously time-consuming method that limits both the patterned device size and the output power. Here, we demonstrate high-power AlGaN-based DUV-LEDs with large-area AlN nanophotonic light-extraction structures that were uniformly fabricated by nanoimprint lithography (NIL),[14,15] thus enabling high-throughput and high-volume fabrication, and a uniform current spreading design that allows high injection current operation. We show that these DUV-LEDs with nanoimprinted AlN nanophotonic structures have a wider near-field emitting area, a stronger far-field extracted light intensity, and an approximately 20-fold increase in output power over that of conventional flat-surface DUV-LEDs. The large-area nanoimprinted single chip DUV-LED produced the record continuous-wave (CW) output power of 150 mW in the UV-C (wavelengths below 280 nm) regime at an injection current of 850 mA with a peak emission wavelength of 265 nm.

The wafers for the AlGaN-based DUV-LEDs were grown by metal-organic chemical vapor deposition (MOCVD) on a 100-μm-thick HVPE-AlN substrate. The MOCVD, HVPE, and LED processing techniques have been described in detail elsewhere.[9,10,13] From bottom to top, the LED structures are composed of a 100-nm-thick AlN homoepitaxial layer, a 1-μm-thick Si-doped n-$Al_{0.75}Ga_{0.25}N$ layer, three multiple quantum well (MQW) active layers, a Mg-doped *p*-AlN electron blocking layer, a *p*-$Al_{0.8}Ga_{0.2}N$ cladding layer, and a *p*-GaN contact layer. We enlarged the emitting area to enhance the output power by reducing the current density. The LEDs were designed to have a uniform current density distribution with a multiple-strip *p*-electrode geometry[12] and were fabricated to have larger active areas (mesa sizes of approximately 0.35 mm^2) than conventional small-area ($\sim$0.1 mm^2) of AlGaN-based LEDs. Ni/Au and Ti/Al/Au vapor-deposited layers were used for the *p*-type and *n*-type contacts, respectively. We identified a light-extraction structure that uses an AlN surface configuration composed of a hybrid structure of two-dimensional (2D) photonic crystals (PhCs) and subwavelength nanostructures[13] as the most suitable candidate nanophotonic light-extraction structure for use with DUV-LEDs on AlN substrates. Figure 1 shows a schematic of the fabrication process flow that was used to construct the large-area AlN nanophotonic light-extraction structure. First, to fabricate a precise, high-aspect-ratio AlN nanophotonic structure, a three-layer resist system consisting of a 480-nm-thick poly(methylmethacrylate)-based sacrificial polymer layer, a 40-nm-thick SiO_2 layer acting as the hard mask, and a 230-nm-thick UV-curable imprint resin layer was coated on the AlN substrate. Second, NIL processing was performed by UV exposure under pressure using a flexible, low-cost polymer-based film mold for soft but uniform contact with the DUV-LED wafer; the mold was fabricated using a high-throughput and repeatable NIL process with a silicon master mold. Third, the imprinted patterns were transferred onto the SiO_2 hard mask using a custom-built inductively

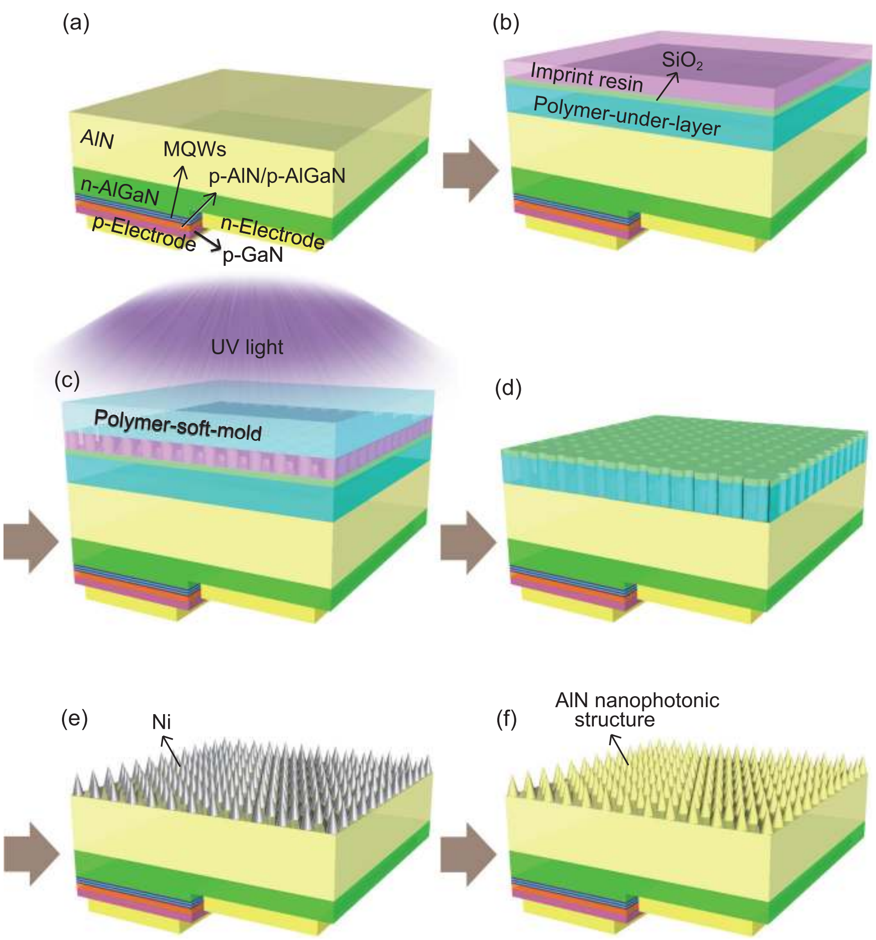

FIG. 1. Schematic diagrams of fabrication process flow used to construct nanophotonic light-extraction structure for DUV-LEDs using soft UV NIL and ICP dry etching processes: (a) after mesa etching and formation of the p- and n-electrodes; (b) the sacrificial polymer underlayer, SiO_2 hard mask, and imprint resin coating; (c) soft UV NIL process; (d) ICP dry etching of the SiO_2 and polymer; (e) Ni deposition and lift-off process; and (f) AlN ICP dry etching and wet etching processes.

coupled plasma (ICP) dry etching system with fluoride-based plasmas, and the sacrificial polymer underlayer was patterned by ICP dry etching using the hard mask pattern and an oxygen-based plasma. Fourth, a Ni hard mask pattern was formed using EB evaporation and lift-off processes. Finally, to construct the AlN 2D PhCs with subwavelength nanostructures on the DUV-LED, the masked AlN surface with the Ni pattern was etched twice, first by ICP dry etching using a fluoride-based plasma and then by wet etching using HCl solutions. The AlN substrate thickness is almost the same before and after the patterning processes. The maximum imprint area is limited by the AlN substrate wafer size and is currently approximately 1 in^2. To enable comparison of the electroluminescence (EL) characteristics of these devices, identical DUV-LEDs with and without the imprinted nanophotonic structures were selected from the same wafer area with the same internal quantum efficiency and the same size and were then flip-chip-mounted on an AlN submount with encapsulation.

Figure 2(a) shows a scanning electron microscopy (SEM) image of the polymer patterns that were imprinted on the SiO_2/polymer sacrificial layer by NIL before the dry etching processes were performed. The image clearly shows that a defect-free periodic polymer pattern was imprinted accurately with a uniform and very thin residual layer. Figure 2(b) shows a cross-sectional SEM image of the Ni arrays formed on the AlN surface after the lift-off process. These high-aspect-ratio Ni circular nanocones were formed using a polymer sacrificial layer with periodic deep holes to fabricate circular AlN cones. Figure 2(c) shows a SEM image of the fabricated AlN nanophotonic light-extraction structure, which is composed of the hybrid structure of 2D PhCs and subwavelength nanostructures. Each 2D PhC was composed of a triangular lattice of circular AlN cones with lattice constant $a = 600$ nm, a filling factor $f \sim 0.9$ at the base of the cones, and an aspect ratio of 1.0. The AlN 2D PhC parameters were determined based on the results of 3D

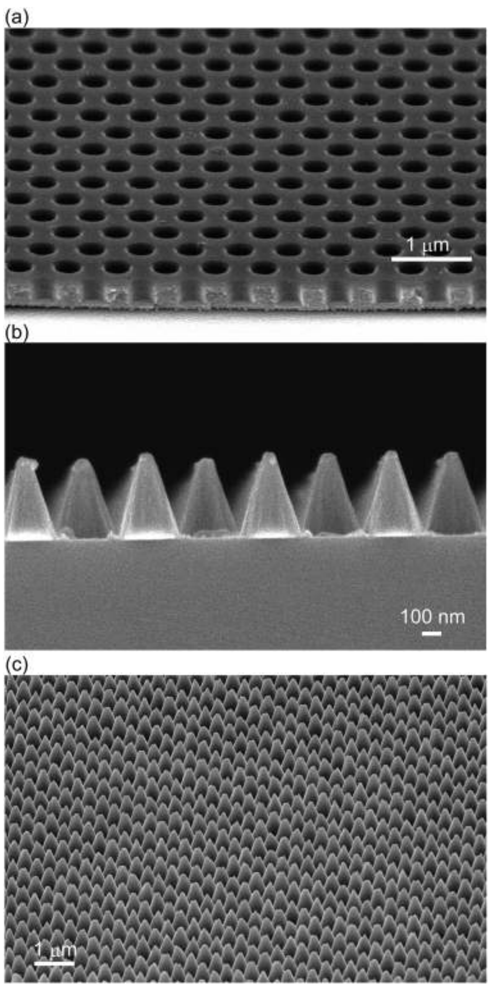

FIG. 2. SEM micrographs of (a) the imprinted resin structure after soft UV NIL, (b) cross-section of Ni arrays on the AlN surface after the lift-off process, and (c) AlN nanophotonic structure fabricated on the light-extraction surface of AlGaN-based DUV-LEDs.

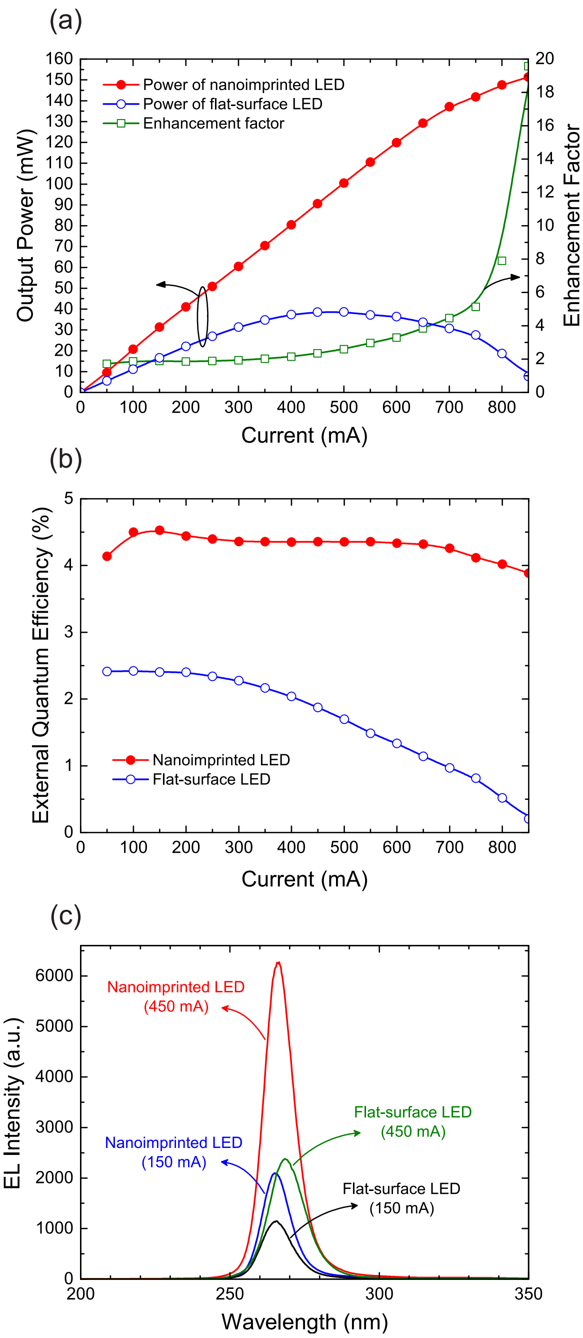

FIG. 3. (a) Output power and enhancement factor characteristics, (b) EQEs as a function of continuous injection current, and (c) EL spectra at two different injection currents of DUV-LED with a nanoimprinted AlN nanophotonic light-extraction structure and a conventional flat-surface DUV-LED.

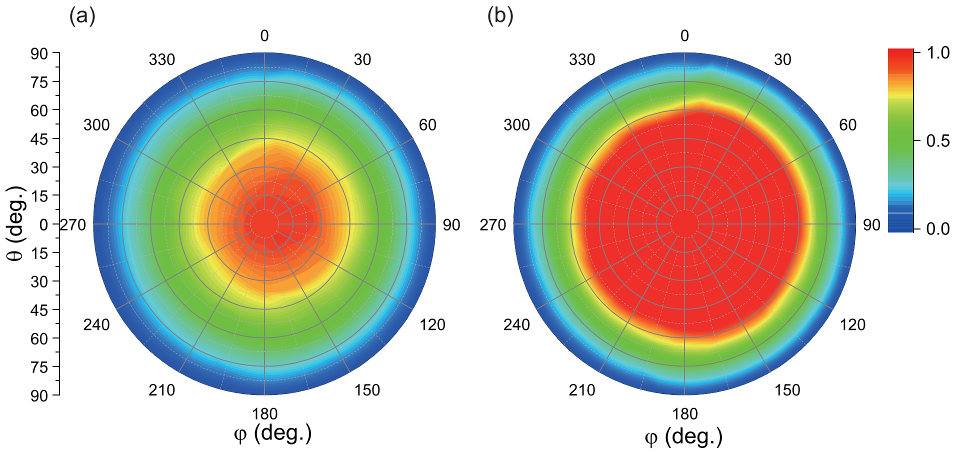

FIG. 4. Full far-field radiation patterns of DUV-LEDs with (a) the conventional flat surface and (b) the AlN nanophotonic light-extraction structure fabricated by NIL.

finite-difference time-domain calculations. Using NIL technology, AlN nanophotonic light-extraction structures were fabricated uniformly on the light-extraction surfaces of the AlGaN-based DUV-LEDs.

Figure 3(a) shows the output powers and the enhancement factor as functions of injection current for the DUV-LEDs with and without the large-area nanoimprinted AlN nanophotonic light-extraction structures. These characteristics were measured under CW conditions at room temperature using a calibrated integrating sphere. The maximum output power of the DUV-LED with the AlN nanophotonic structure was 151 mW at a maximum operating injection current of 850 mA. In contrast, a conventional flat-surface DUV-LED without the AlN nanophotonic structure has a peak output power of 38 mW at 450 mA. The enhancement factor is defined here as the ratio of the output powers of the DUV LEDs with and without the AlN nanophotonic structures. The light extraction enhancement factor of approximately two in the lower injection current region is around 40%–50% higher than reported values in DUV-LEDs that were obtained using conventional random roughening or a moth-eye structure and agrees well with the previously reported enhancement values for small-area LEDs with AlN nanophotonic structures.[13] In contrast, the maximum enhancement factor of 19.6 was observed at a higher injection current of 850 mA. Figure 3(b) shows the EQEs of the DUV-LEDs with and without the AlN nanophotonic structures as a function of injection current. The EQE of the DUV-LED with the AlN nanophotonic structure remains consistently high for higher injection currents when compared with that of conventional flat-surface devices and shows peak values of 4.5% at 150 mA and 3.9% at the maximum operating injection current of 850 mA, indicating a significant improvement in the efficiency droop during high current operation. EL spectra were measured at 150 and 450 mA injection currents for both DUV LEDs, with and without the AlN nanophotonic structures, with results as shown in Fig. 3(c). The same single peak emissions were observed at approximately 265 nm at the lower 150 mA current for both devices. However, a larger spectral redshift was clearly observed at the higher 450 mA current in the flat-surface device when compared with the LEDs with AlN nanophotonic light-extraction structures; this will be discussed later.

Figure 4 shows the far-field radiation patterns of DUV-LEDs (a) without and (b) with the AlN nanophotonic light-extraction structure. These full far-field patterns were measured at an emission angle θ and an azimuthal angle ϕ, where $\theta = 0°$ indicates vertical emission, and $\phi = 0°$ is in the Γ-K lattice direction of a 2D PhC. Clearly, significantly stronger extracted light intensity was observed for the DUV-LED with the AlN nanophotonic structure over the entire angular radiation range when compared with that of the conventional flat-surface LED structure. This is attributed to both the enlarged light escape cone and the reduced Fresnel reflection produced by the large-area AlN nanophotonic structure, which was fabricated using low-cost, mass-production-compatible NIL technology.

Figure 5 shows the near-field light emission patterns that were observed from the top of DUV-LEDs with and without

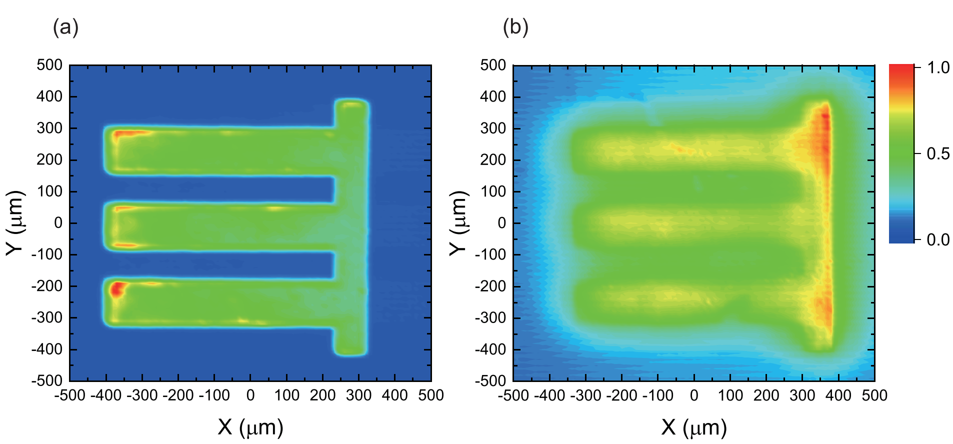

FIG. 5. Near-field light output patterns of 265 nm flip-chip $1 \times 1\,mm^2$ DUV-LEDs with (a) the conventional flat surface and (b) the AlN nanophotonic light-extraction structure.

the large-area AlN nanophotonic light-extraction structure. The full chip dimensions are $1 \times 1\,mm^2$. The *p*-electrode patterns were fabricated for the flip-chip DUV-LEDs with a well-designed multiple-strip *p*-electrode geometry to produce a uniform current density distribution.[12] The near-field pattern of the conventional flat-surface DUV-LED shows uniform DUV emission over the entire *p*-electrode mesa region ($0.35\,mm^2$), which indicates an improvement in the current crowding phenomenon that is commonly observed in AlGaN-based DUV-LEDs with high Al fractions.[4,12] Additionally, the near-field pattern shape of this flat-surface DUV-LED is almost identical to the *p*-electrode mesa pattern, which shows that the DUV light only escapes in the vertical direction through a very narrow escape cone. In contrast, the near-field pattern of the nanoimprinted DUV-LED with the large-area AlN nanophotonic light-extraction structure covering the entire chip area shows not only stronger light intensity than that of the flat-surface DUV-LED but also shows an emitting area that is considerably wider than the mesa region. This is mainly attributed to the enlarged light escape cone that originates from the resonant phenomena that are associated with surface coupling between the leaky radiation modes in the ambient medium and the lateral guided modes in the AlN substrate.[16,17] Additionally, as shown in Fig. 3(c), the EL spectra of the DUV-LEDs with large-area AlN nanophotonic light-extraction structures (which also serve as nanofin structures for heat transfer to the ambient in flip-chip devices) that cover the entire chip area show much smaller spectral redshifts with increasing injection current when compared with those of flat-surface devices, indicating significant suppression of the junction temperature increase in the nanoimprinted LEDs. Here, the junction temperature of the AlGaN-based LEDs can be considered in direct relation to the spectral shift.[18,19] This leads to significant reductions in both optical absorption in and heating of the nanostructured AlN substrate, which is located on the opposite side from the Au/Sn-bonded surface that is in direct contact with the submount. As a result, the DUV-LED with the large-area AlN nanophotonic light-extraction structure shows very low efficiency droop during high injection current operation up to 850 mA.

In conclusion, a high-power single-chip sub-270 nm DUV-LED was demonstrated using a large-area AlN nanophotonic light-extraction structure that was fabricated by NIL. Output power of more than 150 mW was observed at an injection current of 850 mA at a peak emission wavelength of 265 nm. This is the highest reported output power to date for DUV-LEDs with emission wavelengths shorter than 280 nm during CW operation. The LED with the large-area nanoimprinted AlN nanophotonic light-extraction structure shows considerably wider and stronger light emission in its both near-field and far-field patterns, very low efficiency droop, and an approximately 20-fold increase in the output power over that of a conventional flat-surface LED. These high-power DUV-LEDs effectively enable applications where the high power density is required in combination with a compact light source design, bringing substantial advantages over conventional mercury vapor lamps.

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