Milliwatt power UV-A LEDs developed by using n-AlGaN superlattice buffer layers grown on AlN templates

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Abstract
Ultraviolet (UV)-A light-emitting diode (LED) light sources are strongly demanded for both medical and photochemical applications. In our previous report, we investigated the conventional n-AlGaN buffer layer (BL)-based UV-A LED devices and a very low output power was achieved. In this work, we aim for the suppression of vertically propagating threading dislocation densities (TDDs) in the n-AlGaN BL including the current spreading layer (CSL) by introducing Si-doped n-Al\textsubscript{0.37}Ga\textsubscript{0.63}N/n-Al\textsubscript{0.27}Ga\textsubscript{0.73}N superlattices (SLs) between the AlN template and n-AlGaN BL for the demonstration of 341 nm UV-A LEDs. When the conventional n-AlGaN BLs were replaced with n-AlGaN SL-based BLs (with a suitable number of periods up to ~70) in the UV-A multi-quantum wells, then the full width at half maximum of the x-ray rocking curves in the n-AlGaN CSL for the (0002) and (1012) planes, respectively, were reduced to 346 and 431 arcsec and the total TDDs were suppressed to approximately $\sim$1 $\times$ 10\textsuperscript{9} cm\textsuperscript{-2}. Finally, when the conventional Ni (20 nm)/Au (150 nm) p-electrodes were replaced with new Ni (1 nm)/Mg (200 nm) p-electrodes in the n-AlGaN SL-based UV-A LEDs, the maximum output power was improved from 2.1 to 2.5 mW.

Keywords: semiconducting AlGaN, p-AlGaN, low-pressure metalorganic vapor-phase epitaxy, UV-A LED, superlattices, TDDs in n-AlGaN

(Some figures may appear in colour only in the online journal)

1. Introduction

The revolutionary advances in wide bandgap Al\textsubscript{1-x}Ga\textsubscript{x}-N semiconductor materials have attracted much attention since the 1980s, driven by new research demands in the growth process of devices, and strict requirements for high power, low cost, environmental safety and operation in high temperature as well as hardness against high radiation [1–3]. Al\textsubscript{1-x}Ga\textsubscript{x}-N-based ultraviolet (UV)-A light-emitting diode (LED) devices in the problematic wavelength range of 330 – 340 nm have many interesting applications in the areas of sensing, data storage devices, medical, photochemical and materials processing technologies [3–5]. However, the existence of high threading dislocation densities (TDDs) ($\sim$10\textsuperscript{10} cm\textsuperscript{-2}) in the AlGaN buffer layer (BL) grown on sapphire substrate, caused by large lattice mismatches (~13%–16%), have presented a challenge for many researchers [1–6]. In order to resolve this issue of high TDDs in n-AlGaN BL including n-AlGaN current spreading layer (CSL) for UV emission, the crystal structure of AlN template grown on c-(0001)-sapphire substrates was improved...
using a well-known technique of ‘ammonia (NH₃) pulsed-flow multilayer growth’ in Riken [4–6], where full width at half maximum (FWHM) values of the x-ray rocking curves (XRCs) for the (0002) and (10–12) planes, approximately 200 and 350 arcsec, respectively, (TDDs ~5 × 10⁶ cm⁻²) were achieved. But still the growth of AlGaN on BL on AlN template, with x ~ 0.22 Al content for the UV-A range, can have a lattice mismatch >2.2% and can subsequently generate a huge number of vertically propagating TDDs in the subsequent layer of n-AlGaN CSL. Previously, Takano et al demonstrated AlGaN deep ultraviolet (DUV) LEDs grown on high-quality AlN template [4, 5] and achieved a world record external quantum efficiency (EQE) of 20% [7]. In a similar way, Shatalov et al and Kashima et al reported DUV LED devices with EQE of 10% [8, 9], but the performances of DUV LEDs still remain very low compared to blue LEDs [7–12]. Recently, the light extraction efficiencies (LEEs) of DUV LEDs have been improved by both replacing the p-GaN layer with a transparent p-AlGaN layer and the conventional Ni/Au p-electrodes with highly reflective Ni/Al or Rh p-electrodes [4, 5, 7–9, 13–15]. Therefore, we chose the same route of the DUV LED growth for the development of UV-A LEDs grown on AlN template [4–6]. Proper design of the n-AlGaN BL including CSL and multi-quantum well (MQWs) structure was kept in mind for the emission wavelength of the UV-A range, both in term of TDD engineering as well as Al composition for high internal quantum efficiency (IQE) [4–9, 13–15]. To date, very few studies have been performed using AlGaN-based UV-A LED and laser diode (LD) devices grown on AlN templates with emission wavelengths between 320–360 nm. Results from such studies have been reported in [3–5, 16, 17]. Hirayama et al successfully demonstrated the electroluminescence (EL) emission spectrum of 352 nm from quaternary InAlGaN-based MQWs structure, which was grown on AlN template [16]. Later, Hamamatsu Photonics successfully demonstrated AlGaN UV-A LDs with an emission wavelength of 326 nm grown on GaN templates [17]. Yoshida et al demonstrated the lasing action in AlGaN-based UV-A MQWs structure grown on AlGaN and achieved milliwatt power operation with an emission wavelength of 342 nm [18]. In 2016, a 1 W output power at 338 nm emission in a UV-A LD grown on a GaN bulk substrate was successfully achieved by the same group of Hamamatsu Photonics [19]. Yang et al also demonstrated UV-A light emission at 330 nm from AlGaN UV-A MQWs structure [20]. The performances of AlGaN UV LED devices are related to the optimization of the device structure, design parameters of MQWs/ multi-quantum blocking (MQB) and improvements in the epitaxial material quality (TDDs) of AlGaN BLs [3–5, 21]. The device structures of UV-A LED in the problematic wavelength of UV-A emission between 320–350 nm are still very challenging due to the existence of relatively high TDDs ~5 × 10⁶ cm⁻² in the AlGaN BL grown on AlN template and low hole density in the p-AlGaN layer [3, 4, 21]. Marques et al investigated the thermodynamic property of the AlGaN alloys and indicated that most of the nitride alloy systems have strong indications of a miscibility effect. AlGaN alloys are an exception because of the small lattice mismatch between AlN and GaN [22]. There are several promising routes to suppress the problematic TDDs in the n-AlGaN CSL grown on AlN template [3–5, 23]. Among such routes the process is either to introduce superlattice (SL) structure with a suitable number of periods between the AlN and n-AlGaN CSL, or to grow thicker n-AlGaN BL including highly relaxed n-AlGaN CSL (without SLs), with respect to the fully relaxed AlN template. Very recently, our group at Riken reported highly relaxed (75%) n-AlGaN CSL-based UV-A LED structure grown on AlN template, with an emission wavelength of 326 nm without using SLs [23]. In this work, the former route (with SLs) is adopted and the Si-doped alternating n-Al0.37Ga0.63N-In/Al0.25Ga0.75N-II SLs are introduced between the AlN template and n-AlGaN BL, with the goal of the suppression of TDDs in the n-AlGaN CSL. The optimization of SL-based UV-A MQWs structure with a varied number of periods is first investigated to suppress the TDDs in the n-AlGaN CSL crystal before the fabrication of SL-based UV-A LED devices. In this work, both the UV-A light transmittance through p-AlGaN on UV-A-LED as well as reflectance from the new type of p-electrodes are explored.

2. Experimental procedure

In this work, a well-baked low-pressure metalorganic vapor-phase epitaxy reactor was used for the growth of conventional AlGaN-based UV-A LED structure (labeled UV-A LED), n-AlGaN SL-based UV-A MQWs structures (labeled SL UV-A MQWs) and n-AlGaN SL-based UV-A LED structure (labeled SL UV-A LED), respectively, as shown in Figures 1(a)–(c).
Trimethylgallium (TMGa), trimethylaluminium (TMAI), tetraethylsilane (TESi), bis(ethylcyclopentadienyl)magnesium (Cp2Mg) and ammonia (NH3) were used as gas precursors in the reactor. The bubbler temperatures for the TMGa, TMAI, TESi and Cp2Mg gas precursors were set to −4 °C, 17 °C, −10 °C and 40 °C, respectively, under pressure of 760 Torr. The carrier gas flows H2(III) = 2000 sccm, H2 (V) = 1000 sccm, N2(side) = 3000 sccm and N2(top) = 500 sccm, respectively, were supplied to the growth chamber, during the growth of all samples. The TMAI flows were set to 21 sccm for the growth of all samples except the MQWs crystal structure of each sample.

2.1. Crystal growth of conventional n-AlGaN UV-A LED (reference sample-I)

First, the growth of conventional AlGaN-based UV-A LED (sample-I) was carried out, where the growth temperature was set to 1130 °C and NH3 flow was set to 2000 sccm under the reactor pressure of 76 Torr. Following the high quality of 4 μm thick AlN template [4–6], a 1 μm thick Si-doped n-Al0.54Ga0.46N BL and then a Ga-rich 1 μm thick AlN template [4,6] were grown. A 15 nm thick Si-doped n-Al0.25Ga0.75N CSL was grown, using the growth condition of TMGa = 4.0 sccm (for BL), 9.0 sccm (for CSL) and TESi = 0.1 sccm (for both BL and CSL). The Si concentration was kept at the same value of approximately 3 × 1019 cm−3. Next, threefold MQW structure with a 5 nm thick Al,Ga1−xN QW/12 nm thick AlxGaxN quantum well barrier (QWB) were grown, using the growth condition of TMGa = 10.0 sccm for both barriers and wells, TMAI = 4.0 sccm for wells only and 21.0 sccm for barriers only. Subsequently, twofold thin p-Al0.20Ga0.80N blocking (15 nm)/p-Al0.45Ga0.55N valley (10 nm) structures were grown as MQB layers between the MQWs and n-AlGaN cladding layers, using the growth condition of TMGa = 9.0 sccm (blocking) and 4.0 sccm (valley), and Cp2Mg = 30.0 sccm (for both blocking and valley). Finally, a 200 nm thick Mg-doped p-Al0.25Ga0.75N cladding layer including a p-AlGaN contact layer were grown, using the growth condition of TMGa = 9.0 sccm (for p-AlGaN cladding and p-AlGaN contact). Cp2Mg = 70.0 sccm for the p-AlGaN cladding and Cp2Mg = 90.0 sccm for the p-AlGaN contact layer were supplied. The Mg concentration in the p-AlGaN cladding layer was kept around 3 × 1019 cm−3. Figure 1(a) shows the schematic diagram of the conventional UV-A LED structure (sample-I).

2.2. Crystal growth of n-AlGaN SL-based UV-A MQW structure

Next, we grow several samples of the Si-doped n-AlGaN SL-based UV-A MQWs structures by using two alternating layers of Si-doped n-Al0.32Ga0.68N layer-I /n-Al0.27Ga0.73N layer-II SLs grown on AlN template, with a varying number of periods (cycles). The growth temperatures of the SL UV-A MQWs were set to 1140 °C and the NH3 flows were set to 1500 sccm for all samples under the growth pressure of 76 Torr. Briefly, first n-AlGaN SL-based UV-A MQWs sample-A (30 periods), sample-B (50 periods) and sample-C (70 periods), respectively, were grown. They consisted of an approximately 4 μm thick AlN template and approximately 14, 10 and 7 nm thick alternating layers of SL structure (n-Al0.32Ga0.68N layer-I/n-Al0.27Ga0.73N layer-II), respectively. The SL UV-A MQWs sample-A (30 periods), sample-B (50 periods) and sample-C (70 periods) were grown, using the growth condition of TMGa = 5.6 sccm (layer-I) and 9.0 sccm (layer-II), and TESi = 0.1 sccm (for both layer-I and layer-II), with varying periods and growth time condition, as given in Table 1. Next, stacking layers of 1.34 μm thick Si-doped n-Al0.23Ga0.77N BL and 99 nm thick Si-doped n-Al0.18Ga0.82N CSL, were grown on the over layer of the SL structure, using the growth condition of TMGa = 9.0 sccm (BL) and 12.0 sccm (CSL), and TESi = 0.1 sccm (for both BL and CSL). The Si concentration in the n-AlGaN SL, n-AlGaN BL and n-AlGaN CSL layers, respectively, were kept at the same value of approximately 3 × 1019 cm−3. Subsequently, an overlayer of threefold MQW, consisting of approximately 2 nm thick Al0.18Ga0.82N QW and 6 nm thick Al0.23Ga0.77N QWB was deposited on the n-AlGaN CSL, using the growth condition of TMGa = 12.0 sccm (for both QW and QWB), TMAI = 4.0 sccm (QW) and 21.0 sccm (QWB). Finally, an 8 nm thick undoped Al0.23Ga0.77N final barrier (FB) layer was grown on the overlayer of the MQWs structure, using TMGa = 9.0 sccm. Typical schematic structures of the SL UV-A MQWs for sample-A, sample-B, and sample-C are shown in figure 1(b).

2.3. Crystal growth of n-AlGaN SL-based UV-A LED structure

Finally, Si-doped n-AlGaN SL-based UV-A LED structure (sample-II) was grown using the same growth condition of SL UV-A MQWs sample-C, until the FB layer, and then completed the remaining structure with p-AlGaN MQB and p-AlGaN cladding layer including p-AlGaN contact layer. For the growth of MQB and p-AlGaN cladding layer the NH3 flow was changed from 1500 to 2500 sccm. Next, twofold thin p-Al0.18Ga0.82N (20 nm) /p-Al0.18Ga0.82N layers (15 nm) were grown as MQB layers on the similar SL UV-A MQWs structure using the growth condition of TMGa = 7.0 sccm (blocking) and 12.0 sccm (valley), and Cp2Mg = 30.0 sccm (for both blocking and valley). Finally, a 200 nm thick Mg-doped p-Al0.25Ga0.75N cladding layer including a p-AlGaN contact layer were grown, using the growth condition of TMGa = 12.0 sccm (p-AlGaN cladding and p-AlGaN contact), Cp2Mg = 70.0 sccm (p-AlGaN cladding) and 90.0 sccm (p-AlGaN contact). The Mg concentration in the p-AlGaN cladding layer and p-AlGaN contact layer was kept at approximately 5 × 1018 cm−3 and 8 × 1020 cm−3, respectively. A typical schematic structure of the SL UV-A

| Number of periods in the n-AlGaN SLs | Growth time of one period |
|-------------------------------------|--------------------------|
| 30                                  | 15/15 s                  |
| 50                                  | 10/10 s                  |
| 70                                  | 7.5/7.5 s                |
LED structure (sample-II), which is based on sample-C (70 periods) is shown in figure 1(c).

The structural properties including crystallinity, Al composition and strain-relaxation ratio were investigated by XRD measurement of \( \omega \sim 2 \theta \) scan and XRCs as well as reciprocal space mapping (RSM). The TDDs in the n-AlGaN SLs and in the n-AlGaN CSL of the SL UV-A MQWs were investigated by XRCs analysis [24], using an instrument from Malvern Panalytical Japan. Cross-sectional views were conducted with a transmission electron microscope (TEM) by the Foundation for Promotion of Material Science and Technology (MST) of Japan. TEM observation, including a cross-section of high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) images and bright-field STEM images were taken. The surface roughness of the samples was evaluated by atomic force microscopy (AFM). Annealing for the Mg activation in the p-AlGaN layer was conducted in the presence of \( \text{N}_2 \), at 900 °C for 10 min. The photoluminescence (PL), spectra were measured using a 20 mW Ar-SHG (244 nm) laser as an excitation source. Finally, In-dot metal deposition was used for the fabrication of the n-electrode. The output power was measured under bare-wafer conditions using a Si-photodetector, which was located just behind the test samples. It was calibrated to measure the luminous flux for accurate values of output power [14, 15]. The performances of the UV-A LED devices were evaluated at RT under continuous wave (CW) operation using p-type electrodes with chip size area of 0.25 × 0.25 mm².

3. Results and discussions

3.1. Characterization and demonstration of AlGaN-based UV-A LED (without SLs)

The conventional UV-A LED reference sample-I (without SLs) shown in figure 1(a), was successfully demonstrated with an emission wavelength of 342 nm. The desired UV-A PL emission spectra from the MQWs is primarily dependent on the design, crystallinity and optimization of the n-AlGaN BL as well as the n-AlGaN CSL along with suitable MQWs structure [3–5, 21]. The PL spectral emission intensity of the MQWs structure is strongly dependent on the crystalline quality of the MQWs, whereas the crystalline quality of the MQWs is further dependent on the TDDs level in the n-AlGaN CSL. Therefore, the XRCs of the n-AlGaN CSL in reference sample-I were measured first, where the FWHM value of the XRCs for the (10–12) plane was found to be 793 arcsec, as shown in figure 2(a). This can be compared to the AlN template layers used in this work, which were grown on c-plane-(0001)-sapphire substrates with FWHM values of the XRCs for the (0002) and (10–12) planes approximately 200 and 350 arcsec, respectively (TDDs ~ 5 × 10⁵ cm⁻²) [4, 5]. This confirms that the dislocations in the n-AlGaN CSL of sample-I are several times higher than in the AlN template. This shows that the penetration possibility of vertically propagating dislocations, originating from the lattice mismatch at the interface of the AlN template and AlGaN BL, into the n-AlGaN CSL including MQWs of the conventional UV-A LED cannot be ignored.

Figure 2(b) shows the PL emission spectra from MQWs of the UV-A LED (sample-I) in a wide range of wavelength including the n-AlGaN CSL. Single-peak PL emission spectra at 310 nm from the n-AlGaN CSL with relatively high-intensity PL emission spectra at 341 nm from MQWs in the short-wavelength range are demonstrated, as shown in the inset of figure 2(b).

Next, AlGaN-based UV-A LED device performance was evaluated using a conventional Ni (20 nm)/Au (150 nm) p-electrode. The EL emission spectra (EL versus wavelength), current versus output power \((I–L)\), and current versus voltage \((I–V)\) measurements were performed at RT, as shown in figures 2(c) and (d). The tuning of EL emission spectra under different values of dc current was demonstrated, as shown in figure 2(c), and unexpectedly double-peak emission spectra were identified at a 290 and 342 nm peak, respectively, in the UV-A LED (sample-I), which is in contrast to the PL emission spectra of the same sample-I, are shown in figure 2(b). Now, the question arises, whether compositional phase separation was generated or not in the AlGaN alloys due to the internal strain. As known, the compositional phase separation phenomena were not realized in AlGaN alloy materials [22]. Therefore, the double-peak emission spectrum from the AlGaN-based UV-A LED is speculated and regarded to be due to the extended crystal defects that generate localized energy states. Such crystal irregularities cannot be easily observed by TEM or 3D atom probe in the MQWs structure. The possibility of extended defects and crystal irregularities, where the strain relaxation may lead only to the generation of additional dislocations, but can excite 3D growth mode in the AlGaN materials. But the origin of the additional EL peak at 290 nm is still not known and further convincing evidence is still required.

During the \( I–V \) characterization, the driving voltage of 2.5 V at 2 mA was measured, as shown in the inset of figure 2(d). The maximum output power of 1 mW under 98 mA using a Ni (20 nm)/Au (150 nm) p-electrode was demonstrated, as shown in figure 2(d). This low-output power is attributed to both un-optimized n-AlGaN BL including MQWs and a high level of dislocations or extended defects, which is evident from the XRCs observation, as shown in figure 2(a) as well as the double-peak EL emission spectra from MQWs, as shown in figure 2(c). Therefore, an alternative approach of n-AlGaN SL-based BL was proposed in this work to improve the crystallinity of the n-AlGaN CSL beneath the MQWs.

3.2. Electrical and structural characterization of n-AlGaN SL-based UV-A MQWs

In order to improve the performance of UV-A LED, with an emission wavelength of 341 nm, SLs based on alternating layers of n-AlN₀.₃₈Ga₀.₆₂N layer-Inₐ₈Ga₀.₂₂N layer-II with a varied number of periods were introduced between the n-AlGaN BL and AlN template with MQWs structure, as shown in figure 1(b). In this section, the influence of various periods (cycles) in SLs on the surface roughness as well as the emission wavelength range are demonstrated, as shown in the inset of figure 2(b).
Figure 2. (a) XRCs of the n-AlGaN CSL in UV-A LED, where the FWHM value of the \((10\text{-}12)\) plane is shown, and (b) PL emission spectra versus wavelength from the UV-A LED (sample-I), in the wide range of wavelength. (PL emission spectra versus wavelength from the UV-A LED in the short range of wavelength is shown in the inset). Performance evaluation of the conventional n-AlGaN UV-A LED (sample-I), (c) EL emission spectra versus wavelength under different dc drive, and (d) current versus output power, \((I\text{-}V)\) characteristic is shown in the inset).

Figure 3. AFM images of the n-AlGaN SL-based UV-A MQWs, (a) sample-A (30 periods), (b) sample-B (50 periods) and (c) sample-C (70 periods), respectively, and (d) PL emission spectra versus wavelength from n-AlGaN SL-based UV-A MQWs structures of sample-A (30 periods), sample-B (50 periods) and sample-C (70 periods), respectively, at RT.
efficiency of the PL spectral intensities of SL UV-A MQWs sample-A (30 periods), sample-B (50 periods) and sample-C (70 periods), respectively, were investigated using AFM and PL measurement systems at RT, as shown in figures 3(a)–(d). When the number of periods (cycles) in the SLs was varied from 30 periods (sample-A) to 70 periods (sample-C) the root mean square (RMS) values of the FB were decreased from 4.2 to 1.7 nm, respectively, as shown in figures 3(a) and (b). This indicates that the higher the number of periods in SL structure, the smoother the surface of FB in the SL UV-A MQWs will be. The desired PL spectral intensity from the MQWs primarily depends on the crystalline quality of the overall MQWs structure, especially on the QWB thickness and QW thickness. Next, the PL emission spectra of the SL UV-A MQWs in the short range of wavelength for sample-A, sample-B and sample-C, respectively, are investigated, as shown in figure 3(d). Here, the PL spectral intensities from the SL UV-A MQWs were increased in proportion to the number of periods in the SLs. It can be speculated that the dislocations were reduced first in the n-AlGaN CSL and subsequently in the MQWs structure as the number of periods in the SL UV-A MQWs were increased up to 70 periods. The overall crystalline quality of the MQWs structure solely depends on the crystal design of the AlN template, n-AlGaN BL and n-AlGaN CSL, respectively [4, 5, 21].

Table 2. One typical planer RSM cross-section of n-AlGaN SL-based UV-A MQWs structure (sample-C), with mean satellite spacing, ΔQ (rlu) as well as thickness are given.

| (Qx, Qy)−(Qx, Qy) | ΔQ     | Thickness (µm) |
|-------------------|--------|----------------|
| (−0.634 36, 0.792 03)−(−0.634 36, 0.778 64) | 0.01338 | 0.007473 |
| (−0.634 36, 0.778 64)−(−0.634 36, 0.765 32) | 0.01332 | 0.007508 |

Figure 4. (a) XRCs of the n-AlGaN SL-based UV-A MQWs, with the FWHM values of the (0 0 0 2) plane, and (10–12) plane of sample-A, sample-B and sample-C, respectively, and (b) RSM of the n-AlGaN SL-based UV-A MQWs structure (sample-C), along the (−1 −1 4) reflections.

In order to quantify the reduction of dislocations in the n-AlGaN CSL, the crystalline qualities of the SL UV-A MQWs (sample-A, -B and -C), were investigated. The FWHM values of the XRCs in the n-AlGaN CSL for the (0 0 2) and (10–12) planes, respectively, are presented in figure 4(a). When we increased the number of periods from 30 (sample-A) to 70 (sample-C), the FWHM of the XRCs in the n-AlGaN CSLs of the SL UV-A MQWs for the (0 0 2) and (10 2) planes decreased from 429 and 489 arcsec to 346 and 431 arcsec, respectively, as shown in figure 4(a). It was confirmed that the crystallinity in the n-AlGaN CSL of the SL UV-A MQWs structure was reasonably improved using 70 periods in the SL UV-A MQWs (sample-C).

Based on the observation of AFM, PL emission intensities and XRCs, the measurement data of all three sample-A, -B and -C of the SL UV-A MQWs structure, it was found that sample-C is a reasonable choice for the growth and fabrication of SL UV-A LED device structure. However, the precise information of Al composition, TDDs as well as the relaxation status of the n-AlGaN BL and n-AlGaN CSL in the suitable sample-C of SL UV-A MQWs were still unknown. Therefore, we chose sample-C (SL UV-A MQWs) for further investigation using RSM and TEM analysis. The magnified RSM image of the n-AlGaN CSL in the SL UV-A MQWs (sample-C) along the (−1 −1 4) reflection was measured, as shown in figure 4(b). The RSM indicates that the first stacking layer of the Si-doped n-Al0.23Ga0.77N BL is in a highly relaxed state (with a relaxation ratio of 87%) with respect to the fully relaxed AlN template, as shown in the inset of figure 4(b). In a similar way, the Ga-rich n-Al0.18Ga0.82N CSL was found to be highly relaxed too (with a relaxation ratio of 84%) with respect to the fully relaxed AlN template, as shown in the inset of figure 4(b). Such highly relaxed (87%) n-AlGaN BL and highly relaxed (84%) n-AlGaN CSL might be caused by increasing the number of periods to 70 in the SL UV-A MQWs (sample-C). The relaxation ratio up to 90% of n-AlGaN CSL may be explored in future work by further investigating the relationship between the TDDs level and various kinds of SL structure, both in terms of thickness variation as well as compositional variation.
Figure 5. (a) HAADF-STEM images of sample-C taken along the \((1 - 100)\) zone axis, where \(g = [0002]\), (b) bright-field STEM image taken along the \((1 - 100)\) zone axis, where \(g = [11-20]\), (c) magnified bright-field STEM image of point area ‘A1’ taken from figure (b), and (d) magnified HAADF-STEM image of figure (a) around the n-AlGaN SL section area, and (e) magnified HAADF-STEM image of figure (a) around the n-AlGaN CSL/MQWs section area.
The mean satellite spacing of SLs from the planar cross-section of RSM along the (−1 −1 4) reflection of the epitaxially grown SL UV-A MQWs (sample-C) was measured, as given in the table 2. The average thickness of one cycle of SLs was determined to be 7.5 nm, which is in close agreement with the average thickness of one cycle of SLs (~6.1 nm) measured by HAADF-STEM image, as shown in figure 5(d).

The film layer thicknesses and dislocation densities in SL UV-A MQWs structure (sample-C) were evaluated using STEM, as shown in figures 5(a)–(e). In order to see all types of dislocation densities in the AlN template, n-AlGaN SLs and n-AlGaN CSL, respectively, of sample-C the electron incidence direction in the STEM image of figures 5(a) and (b) were set to the (1 1 0 0) zone axis. Figures 5(a) and (b) include both \( g = [0002] \), and \( g = [1 1 2 0] \) vector information, respectively, for the estimation of all types of TDDs. The STEM dark-field image taken along the (1 1 0 0) zone axis for sample-C, is shown in figure 5(b), including the AlN template (part: P1), n-AlGaN SLs (lower part: P2), n-AlGaN SLs (upper part: P3) and n-AlGaN CSL (part: P4), respectively. The total TDDs in AlN part (P1), n-AlGaN SL lower part (P2), n-AlGaN SL upper part (P3) and n-AlGaN CSL (part: P4) of the SL UV-A MQWs, were estimated to be approximately \( 5 \times 10^5 \text{ cm}^{-2} \) [4–6], \( 3.3 \times 10^5 \text{ cm}^{-2} \), \( 2.8 \times 10^5 \text{ cm}^{-2} \) and \( 1 \times 10^5 \text{ cm}^{-2} \), respectively, as given in table 3. The total TDDs in the n-AlGaN CSL were reasonably reduced to \( 1 \times 10^5 \text{ cm}^{-2} \), as shown in table 3. Such reductions of the total TDDs are attributed to both n-AlGaN SL structure with 70 periods (sample-C) as well as G-rich n-AlGaN CSL (84% relaxation ratio), as shown by the RSM observation of figure 4(b). Similar investigation has been carried out by several authors in III–V materials, e.g. AlN/AlGaN SL structure [25]. We also investigated the annihilation mechanism caused by n-AlGaN SLs in the SL UV-A MQWs (sample-C) around the point (A1) taken from the HAADF-STEM image of figure 5(b). The bright-field STEM image of figure 5(c) has been shown at point ‘A1’ has been shown in the bright-field STEM image of the introduction of SLs, as depicted by the dotted line (orange color). The HAADF-STEM images shown in figures 5(b) and (c) can explain very well the main processes occurring during the penetration of TDDs through compressively strained AlGaN-based SL structure. These images clearly demonstrate the TD bending and 90° re-orientation (visible as disappearance of the dislocation), which have been described in different articles since the pioneering work on the filtering effect of SLs in AlGaInN-based LEDs, some of which can be found in the work of Wang et al and some others [26–29]. In particular, one type of vertically propagating TD is first divided into three branches (shaped like a tree) and then eventually bent or annihilated half way through the SL structure in the n-AlGaN SL-based UV-A MQWs, as shown in figure 5(c). Most interestingly, one can see in the lower part of the SL structure horizontally aligned misfit dislocations relaxing the strain, as shown in the figure 5(c). However, there are still several new types of dislocations and agglomeration of dislocations in the same structure. The crystalline quality of the MQWs is strongly dependent on the crystallinity of the underlying n-AlGaN CSL, and therefore it can be speculated that the final dislocation density would be similar to the dislocation density at the end of the n-AlGaN CSL/MQWs or that the real TDDs in the MQWs could be slightly higher than the n-AlGaN CSL.

It is also very important to know about the design parameters of the device i.e. the real thickness of each layer of the device structure, especially of the MQWs structure. The total thicknesses of the n-AlGaN SLs structure, n-AlGaN BL and n-AlGaN CSL, respectively, were measured to be 430 nm and 70 periods. The total thickness of the MQWs structure was also measured and a 30 nm thick MQWs structure was confirmed in the HAADF-STEM image of figure 5(e). The thickness of QW, QB and FB were measured to be 6, 2 and 8 nm, respectively, as shown in the HAADF-STEM image of figure 5(e). It shows that the MQWs region is nicely designed, but the Mg diffusion into the MQWs region from the p-AlGaN region is still unknown. Based on the comprehensive investigation of sample-C, using AFM, XRCs, PL, EL, HAADF-STEM and RSM, sample-C (70 periods) was chosen as a suitable candidate to be used for the growth and characterization of the n-AlGaN SL-based UV-A LED structure.

### 3.3. Demonstration of n-AlGaN SL-based UV-A LED (based on sample-C)

The performance of the newly fabricated SL UV-A LED (sample-II), which was strictly based on sample-C, was evaluated at RT. The LEE of all UV LED devices are strongly dependent on the transparency of the p-AlGaN contact layer as well as the reflectivity from the p-electrodes [30, 31]. In order to avoid UV-A light absorption by using the p-GaN layer structure a highly transparent p-AlGaN layer is inevitably required for the enhancement of LEE in the UV-A LED devices. In this work, we successfully achieved the relative transmittance >80% in the wavelength range from 340–380 nm from the highly transparent p-AlGaN contact layer in the SL UV-A LED device, as shown in figure 6(a). The relative transmittance is defined by the ratio of the light intensity transmitted through a UV-A LED on an AlN/sapphire template divided by that through the AlN/sapphire template only.

| Measurement location Type of TDDs | Dislocation density \( \text{[cm}^{-2} \rangle \) |
|-------------------------------|---------------------|
| n-AlGaN CSL part (P4) | Total dislocation \( 1.1 \times 10^9 \) |
| n-AlGaN SLs Upper part (P3) | Total dislocation \( 2.8 \times 10^9 \) |
| n-AlGaN SLs Lower part (P2) | Total dislocation \( 3.3 \times 10^9 \) |
| AlN Template part (P1) | Total dislocation \( 5 \times 10^8 \) |
Finally, the performance of the SL-based UV-A LED device (sample-II), shown in figure 1(c), was evaluated using both conventional Ni (20 nm)/Au (150 nm) as well as a new type of Ni (1 nm) /Mg (200 nm) p-electrode. The tuning of EL emission spectra under different values of dc current was investigated, where the single-peak emission wavelength remains close to 341 nm in all cases using the Ni/Au p-electrode, as shown in figure 6(b). It was found that the EL spectral intensities increased with increasing the dc drive current from 5 to 30 mA. The double-peak EL emission issue in the former conventional UV-A LED (sample-I), shown in figure 2(c), has been overcome in the SL UV-A LED structure shown in figure 6(b).

The $I$–$V$ characteristic driving voltages of 7 and 10 V at 4 mA, respectively, were identified using both Ni/Au and Ni/Mg p-electrodes (sample-II), as shown in the inset of figure 6(c). However, the turn-on voltages in the SL UV-A LEDs (sample-II) were found to be higher than the conventional UV-A LED (sample-I), as shown in the inset of figure 2(d). These high voltages are attributed to both the bare-wafer level measurement and high resistive p-AlGaN contact layer. The possibility of Mg-atomic diffusion from the electron blocking layer (EBL)/p-ALGaN cladding layer into the MQWs via FB cannot be ignored. The maximum output power and EQE were improved up to 2.13 mW and 0.7%, respectively, in the SL-based UV-A LED by using the Ni/Au p-electrode, as shown in figures 6(c) and (d). In this case, the maximum output power was improved from 1 to 2.13 mW compared to the conventional UV-A LED (sample-I). Due to the low reflectance of the Ni layer (approximately 30%) and Au layer (approximately 34%), the overall LEEs were still found to be lower from the Ni (20 nm)/Au (150 nm) stacking layers in the SL UV-A LED (sample-II). When we replaced the conventional Ni (20 nm)/Au (150 nm) p-electrodes with a new type of Ni (1 nm)/Mg (200 nm) p-electrode in sample-II, the EQE was enhanced from 0.7% to 1% at 45 mA under CW operation at RT, as shown in figure 6(d). Consequently, the maximum output power was also improved from 2.1 to 2.5
mW at 80 mA, as shown in figure 6(c). The milliwatt-level power demonstration of SL UV-A LED grown on AlN template can further be improved by reducing the TDDs in the AlN template through defect engineering in the AlN template using epitaxial lateral overgrowth on nano-PSS or micro-PSS, respectively. In particular, we need high hole density of approximately \((-5 \times 10^{16} \text{ cm}^{-3}\) in the p-AlGaN cladding layer for UV-A emission. Last, but not the least, it is important to deeply investigate the Mg-diffusion mechanism from the EBL/p-AlGaN cladding layer via FB into the UV-A MQWs region as this investigation can further improve the hole injection into the MQWs region.

Summary

Si-doped n-AlGaN SL-based UV-A LED devices were successfully demonstrated with single-peak EL emission spectra of 341 nm. The FWHM of XRCs values of the (0002) and (10–12) planes in the n-AlGaN CSL of SL-based UV-A MQWs were successfully reduced to 340 and 431 arcsec, respectively (total TDDs \(~1 	imes 10^2 \text{ cm}^{-2}\)). When we used the number of 70 periods (cycles) in the n-AlGaN SL-based UV-A LED, the maximal output power of 2.1 mW at 50 mA was achieved. When we replaced the conventional Ni/Au p-electrode with a Ni/Mg p-electrode the EQE was enhanced from 0.7% to 1% at 45 mA under CW operation at RT. Consequently, the maximum output power was also improved from 2.1 to 2.5 mW at 341 nm. The LEE was improved by introducing a highly reflective Ni/Mg p-type electrode and achieving highly transparent p-AlGaN contact layer. The demonstration of UV-A LED is promising for the realization and reinforcement of UV-A LD structure grown on AlN template and it can be a potent device for opening a path to new applications.

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