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Cite as: AIP Advances 9, 125342 (2019); https://doi.org/10.1063/1.5125799
Submitted: 16 October 2019 . Accepted: 05 December 2019 . Published Online: 31 December 2019

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Cite as: AIP Advances 9, 125342 (2019); doi: 10.1063/1.5125799
Submitted: 16 October 2019 • Accepted: 5 December 2019 • Published Online: 31 December 2019

Kanako Shojiki,1,a) Ryota Ishii,2 Kenjiro Uesugi,3 Mitsuru Funato,2 Yoichi Kawakami,2 and Hideto Miyake1,4

AFFILIATIONS
1 Graduate School of Engineering, Mie University, Mie 514-8507, Japan
2 Department of Electronic Science and Engineering, Kyoto University, Kyoto 615-8510, Japan
3 Strategic Planning Office for Regional Revitalization, Mie University, Mie 514-8507, Japan
4 Graduate School of Regional Innovation Studies, Mie University, Mie 514-0001, Japan

a) Electronic mail: k.shojiki@elec.mie-u.ac.jp

ABSTRACT
The impact of a face-to-face annealed sputtered AlN/sapphire (FFA Sp-AlN) template with threading-dislocation densities (TDDs) of 2 × 10^8 cm^-2 and an n-type AlGaN (n-AlGaN) underlayer on optical properties of AlGaN multiple quantum wells (MQWs) with an ultraviolet C (UVC) emission is investigated comprehensively. For comparison of the FFA Sp-AlN template with low TDDs, a conventional MOVPE (metalorganic vapor phase epitaxially)-grown AlN/sapphire (MOVPE-AlN) template with TDDs of 1 × 10^9 cm^-2 was prepared. Consequently, cathodoluminescence (CL), temperature-dependent photoluminescence (PL), and time-resolved PL (TR-PL) measurements verified that both the FFA Sp-AlN template and n-AlGaN underlayer are indispensable for obtaining MQWs with high internal quantum efficiencies, which decrease the TDDs and point defect (PD) densities. Our results revealed that 10-period quantum wells (10QWs)/n-AlGaN/AlN grown on the FFA Sp-AlN template exhibit a lower dark spot density in CL panchromatic intensity maps, a higher integrated emission intensity ratio from the temperature-dependent PL (from 15 to 300 K), and a longer nonradiative lifetime from the TR-PL measurements at 300 K compared with those grown on the MOVPE-AlN template. Moreover, we found that the optical properties of 10QWs/AlN in FFA Sp-AlN and MOVPE-AlN templates do not exhibit a significant difference because of the existence of numerous PDs. Our experimental results demonstrate the favorable impact of the FFA Sp-AlN template for low-TDDs and the n-AlGaN underlayer for low-PDs, which holds promise for highly efficient AlGaN deep-ultraviolet light-emitting devices.

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I. INTRODUCTION

AlGaN based light-emitting devices have developed rapidly1,3 and gained popularity in the medical, environmental, and industrial applications, such as sterilization and water purification systems. For the applications of AlGaN light emitting diodes (LEDs) in the deep ultraviolet (DUV) region, the low-threading-dislocation density (TDD) AlN templates with a large-wafer diameter and low-production cost are highly desirable. The internal quantum efficiency (IQE) of multiple quantum wells (MQWs) generally increases as the TDD decreases.4,5 Usually, sapphire substrates are chosen as the epitaxial substrates for AlN owing to the low-production cost and DUV transparency; however, a large lattice mismatch between AlN and sapphire results in high TDDs. Recently, the face-to-face annealed sputter-deposited AlN (FFA Sp-AlN) template on sapphire has been reported as a viable alternative to the conventional metalorganic vapor phase epitaxially (MOVPE)-grown AlN template (MOVPE-AlN) on sapphire, which forms low-TDD AlN films using low-production-cost sputtering and annealing methods on low-cost sapphire substrates.6,7 In addition to TDDs, point defects (PDs) are
known as one of the major nonradiative recombination centers (NRCs). Indeed, due to the existence of PDs, optical properties independent of TDDs were reported in GaN and InGaN quantum wells (QWs).\(^\text{10}\) On the FFA Sp-AlN templates, the AlGaN LED with the improved external quantum efficiency (EQE) has been reported.\(^\text{11}\) Additionally, the EQE of the AlGaN LED on the FFA Sp-AlN template is comparable to that on the low-TDD epitaxially laterally overgrown AlN/sapphire template.\(^\text{12}\) However, the mechanism of the EQE improvement of LEDs on FFA Sp-AlN templates is not clear.

For AlGaN MQWs, the IQEs are usually estimated by the excitation-power-density-dependent photoluminescence (PL) at room temperature (RT) based on the well-known Shockley-Read-Hall (SRH) recombination,\(^\text{13}\) temperature-dependent PL with an assumption of IQE being 100% at low temperatures, and a combination of both.\(^\text{14\text{–}17}\) Although these methods are convenient and frequently used, the IQE estimation of the AlGaN-MQWs is not simple because of the influence of the saturable SRH recombination centers\(^\text{18\text{–}21}\) and Auger recombination.\(^\text{22}\) Especially, for PL measurements upon a nonselective excitation, it is expected that a carrier/exciton transfer into QWs from the surroundings affects the IQE estimation as it has been reported for the InGaN system.\(^\text{23}\)

In this study, we investigate the optical properties of AlGaN MQWs on the FFA Sp-AlN template to understand the origin of EQE improvements in AlGaN LEDs on FFA Sp-AlN, and we compare the results with that on the MOVPE-AlN. The quantitative evaluation of IQEs of the AlGaN MQWs is still a challenging task because PL measurements upon a selective excitation are difficult in the ultraviolet C (UVC) range. Therefore, instead of a direct evaluation of IQE, we grew samples under the same MOVPE regrowth conditions and compared their optical properties using temperature-dependent PL, time-resolved PL (TR-PL), and cathodoluminescence (CL). As for the MOVPE regrowth structures, in addition to 10 periods quantum wells (10QWs) directly grown on AlN (10QWs/AlN), 10QWs/n-type AlGaN (n-AlGaN)/AlN were grown to assimilate the condition of LED structures and check the impact of n-AlGaN underlayers.

II. EXPERIMENT

Figures 1(a)–1(d) show schematics of the sample structures evaluated in this study. We prepared four sets of samples with 10QWs grown on different templates (FFA Sp-AlN and MOVPE-AlN) on c-plane sapphire substrates. The FFA Sp-AlN template with a thickness of 450 nm was prepared by sputtering and annealing processes.\(^\text{6,7}\) For comparison, the 3-μm-thick MOVPE-AlN template was grown by MOVPE directly on a sapphire substrate. Polarities of both templates were group-III polarity. To investigate the impact of an n-AlGaN underlayer, 10QWs/n-AlGaN/AlN [Figs. 1(c) and 1(d)] were regrown on both the templates along with 10QWs/AlN [Figs. 1(a) and 1(b)]. The substrate off-cut angle of the samples except the 10QWs/n-AlGaN/AlN on the FFA Sp-AlN template is 0.2° toward the m-axis direction of sapphire. For the 10QWs/n-AlGaN/AlN sample on the FFA Sp-AlN template, the c-plane sapphire with an off-cut angle of 0.6° toward the m-axis direction of sapphire is chosen to suppress the hillock formation during the n-AlGaN growth. The impact of the off-cut angle of the sapphire substrate on the n-AlGaN surface morphology is out of scope of this paper and will be discussed elsewhere in detail. The growth chamber pressures for growth of AlN, n-AlGaN1 and n-AlGaN2, and MQWs were 13 kPa, 20 kPa, and 30 kPa, respectively. For the 10QWs/AlN sample, an 800-nm-thick AlN was regrown at a growth temperature \(T_g\) of 1300 °C followed by 10QWs grown at 1050 °C. V/III ratios for quantum barriers and QWs were 4264 and 26 129, respectively. The 10QWs/n-AlGaN/AlN sample consisted of the following regrown layers: 200-nm-thick AlN, 200-nm-thick unintentionally doped (UID) AlGaN layers with an AlN molar fraction of 85% and 75% (UID-AlGaN1 and UID-AlGaN2), 1.5-μm-thick n-AlGaN at a \(T_g\) of 1150 °C (n-AlGaN1), and 70-nm-thick n-AlGaN (n-AlGaN2), and 10QWs under the above-mentioned growth conditions. The design of the quantum well structure is the same for all the samples, consisting of 10 periods of AlGaN (8.4 nm)/AlGaN (2.9 nm). QWs are Si-doped to reduce the NRCs composed of cation vacancies.\(^\text{24,25}\) Si-doping levels in n-AlGaN layers and QWs are approximately in the order of \(10^{18} \text{cm}^{-3}\) and \(10^{17} \text{cm}^{-3}\), respectively. The thicknesses of each QWs and quantum barriers were measured by a cross-sectional transmission electron microscopy (TEM) image of the 10QWs/n-AlGaN/AlN sample with an electron-blocking layer and p-type layers (see the supplementary material, Fig. S1). The AlN molar fractions of AlGaN wells and barriers were 55% and 75%, which were estimated from the combination method of the XRD 2θ-ω profile and cross-sectional TEM image. To study the structural properties of each sample, atomic force microscopy (AFM) images and X-ray diffraction reciprocal space maps (XRD-RSMs) were analyzed.
The FWHMs of the X-ray rocking curves (XRC-FWHMs) of AlN and n-AlGaN at (0002) and (10-12) for all the samples are summarized in Table I. For the XRC measurements at (0002), the incident direction of the X-ray is the sapphire (11-20) direction, which is perpendicular to the substrate-off-cut direction. In addition to XRC-FWHMs, the estimated TDDs for screw and edge components (D_{screw} and D_{edge}) using XRC-FWHMs are also listed in Table I. For AlN samples, the values of D_{screw} and D_{edge} were obtained based on the following well-known equations: 

\[ D_{screw} = \frac{\beta_s^2}{4.35} b_s^2 \quad \text{and} \quad D_{edge} = \frac{\beta_c^2}{4.35} b_c^2, \]

where \( b_s \) and \( b_c \) are Burgers vector \( c \) and \( a/3 \), respectively. We used the lattice constants \( c \) and \( a \) of bulk AlN as 0.498 08 nm and 0.311 11 nm, respectively. \( \beta_s \) is the XRC-FWHM of (0002) (denoted as \( \beta_{(0002)} \)). The value \( \beta_s \) was calculated from \( \beta_{(10\cdot12)} \) and \( \beta_{(00\cdot02)} \) to excerpt edge components with the equation:

\[ \beta_s = \sqrt{\left(\beta_{(10\cdot12)}\right)^2 - \left(\beta_{(00\cdot02)}\right)^2 \cdot \cos^2(x)}/\sin^2(x), \]

where \( x \) is the inclination angle. For n-AlGaN layers, it is difficult to obtain the accurate values of D_{screw} and D_{edge} because of unknown \( b_s \) and \( b_c \). Despite the smaller film thickness of FFA Sp-AlN compared with that of the MOVPE-AlN films, both the FFA Sp-AlN template and n-AlGaN layer on the FFA Sp-AlN template have smaller numbers of TDDs than the MOVPE-AlN template and n-AlGaN layer on the MOVPE-AlN template. Moreover, our surface TEM measurements revealed that TDDs in the FFA Sp-AlN and MOVPE-AlN templates are 2 \( \times \) \( 10^8 \) cm\(^{-2}\) and 1 \( \times \) \( 10^8 \) cm\(^{-2}\), respectively.

For investigation of the optical properties, CL measurements combined with a scanning electron microscope (SEM) at 20 K and 300 K, temperature-dependent PL, and TR-PL at 6 K and 300 K were conducted. SEM-CL measurements were performed upon nonselective excitations. Therefore, we conclude that these hillocks result in the spiral growth that originates from the screw TDs. This difference in the hillock density mainly originated from the differences between the screw- and/or mixed-dislocation densities between FFA Sp-AlN and MOVPE-AlN because noticeable lattice relaxations were not observed in XRD-RSMs (see the supplementary material, Fig. S4). This interpretation agrees with the XRC-FWHM results, i.e., D_{screw} of FFA Sp-AlN is much lower than that of MOVPE-AlN, as shown in Table I. The hillock densities of 8 \( \times \) \( 10^8 \) cm\(^{-2}\) and 1 \( \times \) \( 10^8 \) cm\(^{-2}\) were obtained using the 10 \( \times \) 10 \( \mu \)m\(^2\) AFM images of 10QWs/AlN on FFA Sp-AlN and MOVPE-AlN templates, respectively (see the supplementary material, Fig. S5). The hillock density for 10QWs/AlN on FFA Sp-AlN because noticeable lattice relaxations were not observed in XRD-RSMs (see the supplementary material, Fig. S4). This interpretation agrees with the XRC-FWHM results, i.e., D_{screw} of FFA Sp-AlN is much lower than that of MOVPE-AlN, as shown in Table I. The hillock densities of 8 \( \times \) \( 10^8 \) cm\(^{-2}\) and 1 \( \times \) \( 10^8 \) cm\(^{-2}\) were obtained using the 10 \( \times \) 10 \( \mu \)m\(^2\) AFM images of 10QWs/AlN on FFA Sp-AlN and MOVPE-AlN templates, respectively (see the supplementary material, Fig. S5). The hillock density for 10QWs/AlN on the FFA Sp-AlN template was estimated to be approximately two orders of magnitude lower than that on the MOVPE-AlN template and the relationship was similar to the estimated D_{screw} obtained from XRC-FWHMs. Although, the hillock densities are

### Table I. XRC-FWHMs, estimated TDDs from XRC-FWHMs, and observed TDDs from TEM images of FFA Sp-AlN, MOVPE-AlN templates, and n-AlGaN layers.

|                | XRC FWHMs            | Estimated TDDs (cm\(^{-2}\)) | Observed TDDs (cm\(^{-2}\)) |
|----------------|----------------------|------------------------------|-----------------------------|
|                | (arcsec)             | D_{screw}                    | D_{edge}                    |
| (0002)         | 13                   | 3.6 \( \times \) 10^5        | 2.4 \( \times \) 10^4       | 2 \( \times \) 10^4        |
| (10-12)        | 123                  | 2.4 \( \times \) 10^4        | 1 \( \times \) 10^4         |                            |
| n-AlGaN1 on FFA Sp-AlN on 0.6° off-cut sapphire | 63                   | 3.1 \( \times \) 10^5        | 1.6 \( \times \) 10^5       | 1 \( \times \) 10^4        |
| n-AlGaN1 on MOVPE-AlN on 0.2° off-cut sapphire | 153                  | . . .                        | . . .                       | . . .                      |
| FFA Sp-AlN on 0.2° off-cut sapphire | 120                  | 2.4 \( \times \) 10^4        | 1 \( \times \) 10^4         |                            |
| MOVPE-AlN on 0.2° off-cut sapphire | 300                  | . . .                        | . . .                       | . . .                      |

III. RESULTS AND DISCUSSION

A. Homogeneity of CL peak wavelength

Figures 2(a) and 2(b) show the 5 \( \times \) 5 \( \mu \)m\(^2\) AFM images of 10QWs/AlN on FFA Sp-AlN and MOVPE-AlN templates, respectively. Clearly, the hillock density of 10QWs/AlN on the FFA Sp-AlN template was much lower compared with the structure on the MOVPE-AlN template. The dislocation cores were observed at the top of these hillocks. These morphologies, i.e., the spiral growth initiated by the dislocations with a screw component, were also observed in the AFM images of our 3QWs/AlN (see the supplementary material, Fig. S2). U. Zeimer et al. has also reported the surface morphology with hillocks due to spiral growth. Therefore, we conclude that these hillocks result in the spiral growth that originates from the screw TDs. This difference in the hillock density mainly originated from the differences between the screw- and/or mixed-dislocation densities between FFA Sp-AlN and MOVPE-AlN because noticeable lattice relaxations were not observed in XRD-RSMs (see the supplementary material, Fig. S4). This interpretation agrees with the XRC-FWHM results, i.e., D_{screw} of FFA Sp-AlN is much lower than that of MOVPE-AlN, as shown in Table I. The hillock densities of 8 \( \times \) \( 10^8 \) cm\(^{-2}\) and 1 \( \times \) \( 10^8 \) cm\(^{-2}\) were obtained using the 10 \( \times \) 10 \( \mu \)m\(^2\) AFM images of 10QWs/AlN on FFA Sp-AlN and MOVPE-AlN templates, respectively (see the supplementary material, Fig. S5). The hillock density for 10QWs/AlN on the FFA Sp-AlN template was estimated to be approximately two orders of magnitude lower than that on the MOVPE-AlN template and the relationship was similar to the estimated D_{screw} obtained from XRC-FWHMs. Although, the hillock densities are
approximately one order of magnitude higher than the estimated $D_{\text{screw}}$ obtained from the XRC-FWHMs data. The 10QWs/AlN structure on FFA Sp-AlN has a step-bunched surface. This step-bunched surface is ascribed to a relatively large off-cut angle of 0.6°. Whereas, the structure on MOVPE-AlN exhibits a surface with a large number of hillocks originated from the screw- and/or mixed-dislocation, as shown in Figs. 2(c) and 2(d).

As shown in Figs. 3(a) and 3(b), the CL emission peak wavelength of 10QWs/AlN on FFA Sp-AlN is slightly shorter than that of the structure on MOVPE-AlN, which may be associated with the $T_g$ difference due to the different wafer curvatures during the growth of MQWs. The wafer curvature during the MQW growth for the 10QWs/AlN on the MOVPE-AlN template (thick AlN layer) should be slightly larger than that for the structure on FFA Sp-AlN (thin AlN layer). Therefore, the 10QWs/AlN on the MOVPE-AlN template has slightly-lower MQW $T_g$ and higher Ga-incorporation efficiency than that on the FFA Sp-AlN template. The AFM images and peak wavelength maps show a homogeneous emission for the 10QWs/AlN on the FFA Sp-AlN template with a smaller hillock density. These results are ascribed to the higher Ga incorporation into the side facets of hillocks with dense step edges than the flatter areas.\(^{2,3}\) Figures 3(c) and 3(d) revealed that the homogeneity of the CL peak wavelength of 10QWs/AlGaN/AlN also resembles the surface morphology with the same phenomenon as that of 10QWs/AlN samples. The values of the CL peak wavelength for 10QWs/n-AlGaN/AlN tend to show similar tendency to that of 10QWs/AlN, i.e., the sample on the FFA Sp-AlN template exhibits a shorter peak wavelength than the sample on the MOVPE-AlN template.

### B. Dark spot contrasts and densities in CL panchromatic intensity maps

CL panchromatic intensity maps for 10QWs/AlN and 10QWs/n-AlGaN/AlN are shown in Figs. 4(a)–4(h). The clear dark spot contrasts originated from TDs were not observed in the CL panchromatic intensity maps of 10QWs/AlN on both templates [Figs. 4(a)–4(d)] even at 20 K. Ichikawa et al. proposed that these vague CL panchromatic intensity maps are attributed to the existence of a large number of PDs in $r$-plane Al$_{0.8}$Ga$_{0.2}$N/AlN 3QWs.\(^{34}\) When density of PDs is low enough, the excited carriers diffuse into TDs in the vicinity; hence, the TDs appear as dark spots. However, when PDs are the dominant NRCs, carriers are captured at PDs, and they do not reach to TDs. Therefore, the dark spot contrast originating from the TDs decreases with an increase in the PD density. Based on the above explanation, it can be assumed that the PDs are the dominant NRCs in 10QWs/AlN on both templates. Conversely, 10QWs/n-AlGaN/AlN exhibited clear dark spots originating from TDs on both templates, as shown in Figs. 4(e)–4(h). The dark-spot
contrast increased with a decrease in the measurement temperature. This is owing to the excited carrier diffusion length that depends on both the increase in the diffusion length because of the thermal energy and decrease in the diffusion length because of the activation of PDs. By comparing Figs. 4(e) and 4(f), we observed that the dark spot density of 10QWs/n-AlGaN on the FFA Sp-AlN template was much lower compared with the same structure on the MOVPE-AlN template. The dark spot densities of 10QWs/n-AlGaN on FFA Sp-AlN and MOVPE-AlN templates were obtained to be $2 \times 10^{8}$ cm$^{-2}$ and $6 \times 10^{8}$ cm$^{-2}$, respectively (see the supplementary material, Fig. S6). These densities are approximately the same as those TDDs obtained using the XRC-FWHMs and surface TEM image (as shown in Table I). These clear dark spots originating from TDs prove that the density of PD in 10QWs/n-AlGaN is relatively lower than that of 10QWs/AlN on both templates. The details of these results will be discussed in the following paragraphs.

C. Nonradiative recombination estimated by temperature-dependent PL and TR-PL

Temperature-dependent PL and temporal decay curves of the spectrally integrated TR-PL intensities are shown in Figs. 5(a)–5(e) and 6(a)–6(d), respectively. Table II shows the integral emission intensity ratios at 300 K–15 K ($I_{300K}/I_{15K}$) obtained from the temperature-dependent PL data with the peak excitation power density of 4 MW/cm$^2$. Additionally, the integral emission intensity ratios at 300 K–6 K ($I_{300K}/I_{6K}$) under the excitation conditions same as those of the TR-PL measurements (peak excitation power density being 3 kW/cm$^2$) are shown in Table II. The differences between $I_{300K}/I_{15K}$ (upon 4 MW/cm$^2$ excitation) and $I_{300K}/I_{6K}$ (upon 3 kW/cm$^2$ excitation) indicate the existence of the saturable SRH NRCs. For both 10QWs/AlN and 10QWs/n-AlGaN/AlN, the ratio $I_{300K}/I_{15K}$ on the FFA Sp-AlN template is higher than that on the MOVPE-AlN template. The lifetimes of the fast and slow decay components ($\tau_{fast}$ and $\tau_{slow}$) were obtained by fitting the decay signals from TR-PL measurements with the double exponential equation of $i(t) = A_{fast} \exp(-t/\tau_{fast}) + A_{slow} \exp(-t/\tau_{slow})$, where $A_{fast}$ and $A_{slow}$ are amplitudes of the fast and slow components. Under our experimental conditions, the lifetimes of the samples in TR-PL measurements at 300 K can be regarded as the nonradiative lifetime because the values of $I_{300K}/I_{6K}$ are below 50% (Table II). Our TR-PL measurements of the 10QWs/AlN on both templates at 300 K revealed that there were no obvious differences in the lifetimes of both the samples; therefore, no noticeable reduction in the
nonradiative lifetime of 10QWs/AlN was observed even on the FFA Sp-AlN templates, which possess lower TDDs. These results indicate that the high density of PDs in the 10QWs/AlN sample may hinder the impact of TDDs on the templates. The TR-PL results of the 10QWs/n-AlGaN/AlN at 300 K are significantly different from those of the 10QWs/AlN; TR-PL decays of the 10QWs/n-AlGaN/AlN at 300 K, indicating the impact of underlying templates. We observed that 10QWs/n-AlGaN/AlN on the FFA Sp-AlN

![FIG. 5. Temperature-dependent PL of 10QWs/AlN on (a) FFA Sp-AlN and (b) MOVPE-AlN templates; and 10QWs/n-AlGaN/AlN on (c) FFA Sp-AlN and (d) MOVPE-AlN templates with an excitation peak power density of 4 MW/cm². (e) Integrated intensity as a function of the measured temperature.](image-url)

![FIG. 6. TR-PL decay signals of 10QWs/AlN at (a) 6 K and (b) 300 K; and 10QWs/n-AlGaN/AlN on (c) FFA Sp-AlN and (d) MOVPE-AlN templates at 300 K.](image-url)
template has a longer nonradiative lifetime for both $\tau_{\text{fast}}$ and $\tau_{\text{slow}}$ at 300 K; whereas, the lifetimes observed at low temperature (6 K) are almost identical for 10QWs/n-AlGaN/AlN on both the templates. Thus far, we could not conclude that the lifetimes observed at low temperature (6 K) represent radiative lifetimes. This is because the PDs existed as the dominant NRC even at low temperature, especially in 10QWs/AlN, which could be deduced from the CL panchromatic intensity maps [Figs. 4(a) and 4(b)]. In the case of 10QWs/n-AlGaN/AlN, a decrease in the concentration of NRCs with decreasing TDD was ascertained by an increase in the nonradiative lifetime at 300 K. By changing the template from MOVPE-AlN to FFA Sp-AlN, the values of $\tau_{\text{fast,300K}}$ and $\tau_{\text{fast,100K}}$ for 10QWs/n-AlGaN/AlN increased from 2.7 ns and 8.0 ns to 3.8 ns and 19.6 ns, respectively. It should be noted that any enhancements in the localized emission owing to screw dislocation could not be observed with the FFA Sp-AlN template because the screw- and/or mixed-TDD was low enough to avoid spiral hillock formations, as shown in Figs. 2(a) and 2(c). In this study, the lifetimes observed at both low temperature and RT are longer than those reported in the literature.\(^{3,29,33}\)

The longer lifetimes may be attributed to the large quantum-confined Stark effect owing to thick QWs and barriers because the carrier lifetime strongly depends on the structure of the quantum well. It is well known that the thick QWs and barriers exhibit a longer radiative lifetime at low temperatures in the case of GaN/AlGaN QWs and AlGaN/AlN QWs.\(^{31,32}\) On the other hand, the longer lifetime at 300 K indicates the high-crystalline quality of our samples.

**D. Impact of n-AlGaN layers**

Besides the impact of TDDs on IQE, our results from the temperature-dependent PL, TR-PL, and CL measurements indicated that the IQE strongly depends upon the existence of an n-AlGaN underlayer. For example, the 10QWs/AlN contained numerous PDs; therefore, we could not observe the dark spots originating from TDDs in the CL maps [Figs. 4(a)–4(d)]. In contrast, the dark spots were clearly observed for the 10QWs/n-AlGaN/AlN [Figs. 4(e)–4(h)]. The absence of the dark-spot contrasts in CL panchromatic intensity maps of 10QWs/AlN [Figs. 4(a)–4(d)] indicates a shorter carrier diffusion length and a shorter excited carrier lifetime. Our TR-PL results show a good agreement with this explanation: the carrier lifetimes (both, $\tau_{\text{fast}}$ and $\tau_{\text{slow}}$) obtained at 300 K for 10QWs/AlN are shorter than that of 10QWs/n-AlGaN/AlN on both templates. This difference is more pronounced on the FFA Sp-AlN template with low TDDs. An AlGaN/GaN superlattice interlayer in the cubic GaN system\(^2\) and an indium containing interlayer in the hexagonal InGaN system are reported to effectively reduce PDs.\(^{34–43}\) The mechanism for suppressing PDs in AlGaN MQWs is beyond the scope of this paper. Thus far, our empirical results and previous literature\(^{34–43}\) imply that inserting an n-AlGaN layer causes a significant reduction in the PD density, which improves the IQE of the 10QWs/n-AlGaN/AlN on the FFA Sp-AlN template (low-TDD structure). In addition, the absolute intensities (e.g., $I_{300K}$) of 10QWs/n-AlGaN/AlN were observed to be higher than those of 10QWs/AlN [Fig. 5(e)]. As a result, the intensity ratio $I_{300K}/I_{15K}$ of 10QWs/AlN was higher than that of 10QWs/n-AlGaN/AlN, as shown in Table II. These results strongly suggest that the IQE of 10QWs/AlN is not 100% at low temperature, and the IQE of 10QWs/AlN is lower than that of the 10QWs/n-AlGaN/AlN at RT. By comparing temperature-dependent PL spectra of each sample [Figs. 5(a)–5(d)], one could notice that spectra line widths of 10QWs/n-AlGaN/AlN [Figs. 5(c) and 5(d)] are larger than those of 10QWs/AlN [Figs. 5(a) and 5(b)]. This might be due to larger compositional and thickness fluctuations caused by thick n-AlGaN layers, as shown in Figs. 5(a)–5(d). These fluctuations in 10QWs/n-AlGaN/AlN might hinder the clear blue shift with increasing measurement temperature, which could be observed in 10QWs/AlN.

**E. Mechanism of EQE improvement by using FFA Sp-AlN**

As discussed in the introduction section, an exact estimation of IQEs is difficult because our PL measurements were performed under nonselective excitation conditions. Nevertheless, the comprehensive results obtained in this study revealed the IQE improvement of MQWs by using the FFA Sp-AlN template. Both the FFA Sp-AlN template and n-AlGaN underlayer are indispensable for obtaining high IQEs, which decrease the TDDs and PD densities, respectively.

One other candidate that affects IQEs of AlGaN MQWs is the substrate off-cut angle. As described in Sec. II, in this study, to suppress the formation of large hillocks, a 10QWs/n-AlGaN/AlN structure on FFA Sp-AlN was grown on sapphire with an off-cut angle of 0.6°, whereas that on MOVPE-AlN was grown on an off-cut angle of 0.2°. There are several reports that discuss the effect of the substrate off-cut angle and the resulting step-bunched morphology on the properties of DUV-LEDs and AlGaN MQWs.\(^{30–34,44–51}\) The sapphire-substrate off-cut angle around 1.0° is often regarded.

**TABLE II. Summary of temperature-dependent PL and TR-PL results.**

| Temperature dependent PL | TR-PL decay (ns) | (3 kW/cm²) |
|--------------------------|-----------------|------------|
|                          | $I_{300K}/I_{15K}$ | $I_{300K}/I_{4K}$ | $\tau_{\text{fast}}$ | $\tau_{\text{slow}}$ | $\tau_{\text{fast}}$ | $\tau_{\text{slow}}$ |
| 10QWs/AlN               | On FFA Sp-AlN   | 0.63        | 0.03        | 9.5      | 39.8     | 2.7      | 6.4     |
|                         | On MOVPE-AlN    | 0.51        | 0.07        | 7.9      | 38.1     | 2.4      | 6.6     |
| 10QWs/n-AlGaN/AlN      | On FFA Sp-AlN   | 0.42        | 0.20        | 8.6      | 37.5     | 3.8      | 19.6    |
|                         | On MOVPE-AlN    | 0.30        | 0.08        | 9.1      | 34.8     | 2.7      | 8.0     |
as an optimum angle to reduce TDDs and to enhance carrier localization.\textsuperscript{9,29,34} The elucidation of the effect of the substrate off-cut angle on the properties of $n$-AlGaN/FFA Sp-AlN is out of scope of this paper and will be discussed elsewhere.\textsuperscript{10} Nevertheless, we should note that the reduction of TDDs by using FFA Sp-AlN has much more pronounced effects than the introduction of the substrate large off-cut angle. For example, M. Kaneda \textit{et al.} reported the reduction of both AlN (10-10) XRC-FWHMs and estimated TDDs to 638 arcsec and $5 \times 10^4$ cm$^{-2}$, respectively, by using the sapphire substrate with the off-cut angle of 1.0$^\circ$.\textsuperscript{49,50} Similarly, the reduction of XRC-FWHMs of Al$_{0.99}$Ga$_{0.01}$N (10-12) from around 550 arcsec to around 400 arcsec by increasing the off-cut angle from 0.2$^\circ$ to 1.0$^\circ$ was reported by Kojima \textit{et al.} Meanwhile, as shown in Table I, the FFA Sp-AlN possessed the XRC-FWHM of AlN (10-12) of 123 arcsec and the observed TDD of $2 \times 10^4$ cm$^{-2}$ even on the sapphire substrate with an off-cut angle of 0.2$^\circ$. Therefore, TDDs of FFA Sp-AlN on the 0.2$^\circ$ off-sapphire substrate are clearly lower than those of conventional MOVPE-AlN on the sapphire substrate with the larger off-cut angle such as 1.0$^\circ$. As for carrier localization, 10QWs/$n$-AlGaN/AlN on both FFA Sp-AlN and MOVPE-AlN exhibited the carrier localizations with similar scale owing to the step-bunching and hillock formations, as shown in Figs. 3(c) and 3(d). In addition, 10QWs/$n$-AlGaN/AlN on FFA Sp-AlN on the 0.6$^\circ$ off-sapphire substrate has the shorter PL peak wavelength than that on MOVPE-AlN on the 0.2$^\circ$ off-sapphire substrate, as shown in Figs. 3(c) and 3(d). Moreover, the emission intensity improvement owing to the carrier localization could not be observed in CL panchromatic images as shown in Figs. 4(e) and 4(f). Therefore, the improvement of emission intensity by the carrier localization owing to the step-bunch morphed resulting from large substrate off-cut angle is not likely at least within the range of our results in this study.

Thereby, we could finally conclude that the FFA Sp-AlN with low TDDs is favorable to obtain MQWs/$n$-AlGaN/AlN with high IQE. As a result, an improvement of EQE of AlGaN LEDs with MQWs/$n$-AlGaN/AlN on FFA Sp-AlN template with high IQE was ascertained.

IV. CONCLUSIONS
We demonstrated the IQE improvement of 10QWs/$n$-AlGaN/AlN on the FFA Sp-AlN template using the results of CL, temperature-dependent PL, and TR-PL measurements. Our 10QWs/$n$-AlGaN/AlN on the FFA Sp-AlN template can be directly applied to DUV-LEDs. In the CL panchromatic intensity maps, we observed smaller dark spot density associated with TDs in 10QWs/$n$-AlGaN/AlN grown on the FFA Sp-AlN template rather than the same structure grown on the MOVPE-AlN template. Our PL and TR-PL results showed that 10QWs/$n$-AlGaN/AlN possesses a higher $I_{10K}/I_{300K}$ ratio and a longer carrier lifetime at 300 K than the sample grown on the MOVPE-AlN template. Additionally, the 10QWs/AlN sample possesses high PD densities, which hinders the impact of TDDs on the IQE of MQWs. Moreover, AFM images and CL peak wavelength maps revealed that the MQWs grown on the FFA Sp-AlN template exhibited smooth surfaces with step-flow growths and homogeneous emissions with smaller compositional and structural (thickness) fluctuations. These optical properties of 10QWs/$n$-AlGaN/AlN grown on the FFA Sp-AlN template hold promise for the realization of highly efficient DUV-LEDs.

SUPPLEMENTARY MATERIAL
See the supplementary material for the cross-sectional TEM image of 10QWs, AFM images of 3QWs/AlN, AFM images after KOH etching, XRD-RSMs, 10$\times$10 $\mu$m$^2$ AFM images, and dark spot densities from CL panchromatic intensity maps of 10QWs/$n$-AlGaN/AlN.

ACKNOWLEDGMENTS
This work was partially supported by the MEXT “Program for Building Regional Innovation Ecosystems,” the MEXT “Program for Research and Development of Next-Generation Semiconductor to Realize Energy-Saving Society,” JSPS KAKENHI (Grant Nos. 16H04180H, PI17H06762, and 19K15025), JST CREST (Grant No. 16H15710), JST SICORP EU, and the Consortium for GaN Research and Applications.

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