Improved emission intensity of UVC-LEDs from using strain relaxation layer on sputter-annealed AlN

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This paper presents the effects of Mg concentration in a Mg-doped aluminum nitride (AlN) strain relaxation layer according to the metalorganic vapor-phase epitaxy method. A UV LED using a Mg-doped AlN strain relaxation layer at a Mg concentration of $3 \times 10^{19}$ cm$^{-3}$ on a sputter-annealed AlN template produced light output power 11 times as high as that with conventional LED structures. The AlGaN-on-AlN relaxation rate and LED light output power increased starting from Mg concentrations of $10^{18}$ cm$^{-3}$. These characteristics had almost the same values when the Mg concentration was less than $3 \times 10^{19}$ cm$^{-3}$. These results show the improvement of efficiency caused by void formation due to the inversion domain.

1. Introduction

Recently, UVC-LEDs have attracted much attention owing to their use in many kinds of applications such as sterilization of water, air purification, and biomedical applications.1–4) To realize high-performance UVC-LEDs, growth methods of high-crystalline-quality aluminum nitride (AlN) such as AlN substrates and sputter-annealed AlN have been investigated.5–9) However, the external quantum efficiency of UVC-LEDs using these methods is limited to as low as a few percent at a wavelength of strong bactericidal effect.10–15) Dislocations in the LED layers are some of the limiting factors.16–18) For example, a UVC-LED in 265 nm has Al composition of 60%–75% in n-type AlGaN, and the theoretical relaxation critical thickness is less than 100 nm. As a result, this increases the dislocation density in the AlGaN active layer and the internal quantum efficiency becomes low. Conversely, stress in the active region becomes high in the case of coherent growth using an AlN underlying layer. Therefore, the potential height in the electron blocking layer is decreased due to the strong piezoelectric field.19–21) The phenomenon shows that injection efficiency in UVC-LEDs is difficult to improve without stress relaxation. Consequently, the AlGaN active layer in UVC-LEDs requires not only decreasing dislocation density but also decreasing stress between AlN and AlGaN.

The Mg doping technique is commonly used for growing p-type layers. Thus, the acceptor level rises to close the valence band edge due to increasing Mg concentration.22–25) However, Mg concentrations exceeding $3 \times 10^{19}$ cm$^{-3}$ generate voids in AlGaN due to the inversion domain and defects including voids are compensated by acceptors as well as donors.26–28) This void formation by Mg doping is known to have a bad effect on p-type AlGaN.

Herein, we report a strain relaxation effect on an AlGaN layer grown on an AlN template using the Mg doping technique. It is assumed that the generated voids by excess Mg doping contribute to strain relaxation. Because of this effect, we can say that a remarkable improvement in the emission intensity of UVC-LEDs has been achieved.

2. Experimental methods

We grew AlGaN underlying layers with/without strain relaxation layers on sputter-annealed AlN/sapphire templates. These layers were grown using metalorganic vapor-phase epitaxy (MOVPE; Taiyo Nippon Sanso SR–4000HT). Sputter-annealing AlN with face-to-face annealing is one of the fabrication methods that produce AlN of the highest crystalline quality.5) The surface morphology of AlN was similar to that of bulk AlN crystal on the sapphire substrate. Figure 1(a) shows an AFM image of AlN after homo-epitaxial growth using MOVPE while Fig. 1(b) shows a conventional AlN with a low-temperature buffer layer. Both samples were observed to have clear atomic steps; however, spiral growth was confirmed due to a screw dislocation effect when sputter-annealed AlN was not used as shown in Fig. 1(b). It was observed that a very smooth surface was obtained when sputter-annealed AlN was used as shown in Fig. 1(a). This is because the screw dislocation was very low at a screw dislocation density of less than $1 \times 10^6$ cm$^{-2}$. The FWHMs of the XRD rocking curves of sputter-annealed AlN and conventional AlN were 45 and 200 arcsec (0002) and 280 and 400 arcsec (10–12), respectively. We used these AlN templates in our experiment. For comparison, AlN of a total thickness of 3 μm was used in the sputter-annealed AlN and 170 nm was used for the conventional AlN.

The growth temperatures of the AlN, Mg-doped AlN, and AlGaN were 1340 °C, 1025 °C, and 1150 °C, respectively. The low growth temperature for the Mg–AlN layer facilitated effective Mg incorporation into the AlN. Trimethylgallium, trimethylaluminum and ammonia (NH3) were used as precursors.

Figure 2 shows the structure of the UVC-LEDs. Mg doping concentration in the strain relaxation layer was controlled by the flow rate of Bis cyclopentadienyl magnesium and the Mg

Fig. 1. (Color online) AFM images of AlN after MOVPE growth at thickness of 3 μm: (a) sputter-annealed AlN and (b) MOVPE AlN.
concentration was determined using secondary-ion mass spectrometry. The electron blocking layer was unintentionally doped (UID) AlN with a thickness of 20 nm.\textsuperscript{20,29,30} The thickness of the grown n-Al\textsubscript{0.6}Ga\textsubscript{0.4}N and p-Al\textsubscript{0.6}Ga\textsubscript{0.4}N was 2 μm and 20 nm, respectively. After growth for the fabrication of LEDs, the n-type contact region was etched down up to the n-AlGaN cladding layer using Cl\textsubscript{2} plasma reactive-ion etching. The n- and p-type contacts were formed using EB-deposited indium tin oxide. The relaxation ratio and the molar function of AlN were studied using X-ray reciprocal lattice space mapping (RSM).

3. Results and discussion

3.1. Strain relaxation and light output power in UVC-LEDs depend on Mg concentration

The strain in AlGaN and AlN growth is possibly relaxation strain generated due to planar dislocation as a result of superlattice and void formation by epitaxial lateral overgrowth.\textsuperscript{31–34} However, it is difficult to control the relaxation ratio when using these methods. The void defects formed due to excess Mg doping are generated when the Mg concentration is higher than \(3 \times 10^{19} \text{ cm}^{-3}\) at a stable state.\textsuperscript{25–27} We show a causal relationship between void forming and strain relaxation and how the strain relaxation ratio is influenced by Mg concentration. In this section, the relationship between the strain relaxation ratio and light output power in LEDs is investigated at different concentrations of Mg in the strain relaxation layer.

Figure 3 shows an X-ray RSM of the (−105) reflection obtained at Mg concentrations of \(3 \times 10^{20}, 1 \times 10^{20}, 3 \times 10^{19},\) and \(1 \times 10^{19} \text{ cm}^{-3}\). Certainly, the \(a\)-axis lattice constant of AlN was comparatively close to the bulk AlN value; therefore, the AlN was relaxed against the sapphire. The peak of the UID-Al\textsubscript{0.8}Ga\textsubscript{0.2}N layer was moved to the right side when the Mg concentration of the strain relaxation layer was higher than \(3 \times 10^{19} \text{ cm}^{-3}\) and the n-type AlGaN peak became slightly sharp with Mg content of \(3 \times 10^{20} \text{ cm}^{-3}\). The n-AlGaN crystalline quality by the FWHM of the XRD rocking curves with Mg concentration of \(3 \times 10^{20} \text{ cm}^{-3}\) and \(1 \times 10^{19} \text{ cm}^{-3}\) was 310 and 400 arcsec (0002) and 520 and 550 arcsec (10–12), respectively.

Figure 4 shows the strain relaxation ratio of AlGaN as a function of Mg concentration from the result in Fig.3. The AlGaN layer of both the Al\textsubscript{0.8}Ga\textsubscript{0.2}N buffer and Al\textsubscript{0.6}Ga\textsubscript{0.4}N was relaxed from a Mg concentration of \(3 \times 10^{19} \text{ cm}^{-3}\). The result indicates that the Al\textsubscript{0.8}Ga\textsubscript{0.2}N buffer on the strain relaxation layer determined the relaxation ratio and Al\textsubscript{0.6}Ga\textsubscript{0.4}N including n-type AlGaN led to this effect of relaxation. The relaxation ratio of the UID-AlGaN of LEDs with and without a Mg-doped strain relaxation layer was estimated to be 11.7% and 5.9%, respectively. The relaxation ratio of the n-AlGaN layer of LEDs with and without a Mg-doped strain relaxation layer was estimated to be 27.3% and 23.3%, respectively. These results show that both strain relaxation and improvement of crystalline quality are realized by Mg doping.

The behavior of threading dislocation in the LED was measured both with and without Mg doping in the strain
relaxation layer using a transmission electron microscope (TEM) as shown in Fig. 5. The dislocation density in the TEM image can be used to compare each layer with the underlying AlN layer because the measurement sample thicknesses were different (but under the same AlN growth conditions). The dislocation density of n-AlGaN in samples (a) and (b) was 0.875 times and 1.1 times the dislocation density of the corresponding AlN underlying layer. Therefore, the dislocation density of n-AlGaN without a Mg–AlN layer was greater than that of the AlN underlayer. However, in the case of the Mg-doped strain relaxation layer, the n-AlGaN dislocation density was lower than that of the AlN underlayer.

Figure 6 shows that the light output power of the UVC-LEDs depended on the Mg concentration. Light output power improved starting from a Mg concentration of $3 \times 10^{19}$ cm$^{-3}$. This is similar to the strain relaxation ratio of UID-AlGaN as a function of Mg concentration as shown in Figs. 3 and 4. The strain relaxation ratio in UID-AlGaN also became high from a Mg concentration of $3 \times 10^{19}$ cm$^{-3}$ upward. These phenomena suggest that the void formation due to excess Mg doping was caused by strain relaxation and LED efficiency improved with strain relaxation in the device structure. The improvement in efficiency of the LEDs cannot be explained in detail because the difference in dislocation densities in AlGaN with and without Mg doping is too small as shown in Fig. 5. However, it is found that Mg doping and strain relaxation are effective methods to improve the efficiency of UVC-LEDs.

### 3.2. UVC-LED on sputter-annealed AlN template

For comparison, we fabricated UVC-LEDs on conventional AlN with a low-temperature AlN buffer layer as shown in Fig. 1(b). The difference of these samples is in their surface morphology as well as their crystalline quality from XRD FWHMs. Figure 7 shows the EL spectra of UVC-LEDs on conventional AlN and on sputter-annealed AlN with and without Mg doping in the strain relaxation layer at 100 mA injection. The Mg concentration of the strain relaxation layer was $3 \times 10^{20}$ cm$^{-3}$ in the Mg-doped sample. For the LED on conventional AlN doped with Mg, the intensity of the spectrum is 7.5 times as high as that for the non-Mg-doped sample. It is obvious that doping the strain relaxation layer with Mg is an effective way of improving the efficiency of a UVC-LED. Without Mg doping, the samples of LED on sputter-annealed AlN and on conventional AlN did not show different intensities. The reason is that many new dislocations were generated at the AlGaN and AlN interface. The spectral intensity of the LED with sputter-annealed AlN is 1.5 times as high as that with conventional AlN. The result shows the effects of
different crystalline quality in AlN in terms of the strain relaxation generated.

4. Conclusions
We have demonstrated the remarkable effect of a highly Mg-doped strain relaxation layer on the emission intensity of deep-UV (DUV) AlGaN-based LEDs. We have found that the self-formed voids due to excessive Mg doping are very effective in increasing strain relaxation and lead to a remarkable improvement of the efficiency of output. The strain relaxation ratio in the UID-AlGaN layer doubled when a strain relaxation layer was introduced and the improvement of crystalline quality was also confirmed using XRD analysis. We have confirmed that the emission intensity of LEDs with a strain relaxation layer on sputter-annealed AlN is 11 times as high as when a strain relaxation layer is not used. Using the strain relaxation layer due to excessive Mg doping is a more effective method of growing crystals for DUV LEDs.

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Fig. 7. (Color online) EL spectra of several LEDs at injected current of 100 mA. Red: LED on sputter-annealed AlN with Mg doping in strain relaxation layer. The Mg concentration in the strain relaxation layer was $3 \times 10^{20}$ cm$^{-3}$. Gray: LED on conventional AlN with Mg doping in strain relaxation layer.

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