Investigate the Double Peaks in Main Emission of UVB LEDs

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Abstract

In this study we suppressed the parasitic emission caused by electron overflow found in typical UVB light-emitting diodes (LEDs). Furthermore, modulation of the p-layer structure and doping profile allowed us to decrease the relaxation time of the holes to reach conditions of quasi-charge neutrality in the UVB quantum well. Our UVB LED (sample A) exhibited a clear exciton emission, with its peak near 306 nm and a band-to-band emission at 303 nm. The relative intensity of the exciton emission of sample A decreased as a result of a thermal energy effect. At temperatures of up to 363 K, sample A displayed the exciton emission. Our corresponding UVC LED (sample B) exhibited only a Gaussian peak emission at a wavelength of approximately 272 nm.

Introduction

Compact high-efficiency solid-state ultraviolet (UV) light sources, including light-emitting diodes (LEDs) and laser diodes, are of considerable technological interest for use as alternatives to large, toxic, low-efficiency gas lasers and mercury lamps. In addition, mercury lamps are common sources of UV radiation, particularly at shorter wavelengths, and they are bulky and require high voltages for operation. LEDs as replacements would be attractive in terms of their smaller size and lower operating voltages. [1] There are several definitions of UV bands depending on the concern, application, or industry. For health and safety purposes, UV light is commonly divided into UVA (400–320 nm), UVB (320–280 nm), and UVC (280–100 nm) bands. [2, 3] UV light can have damaging effects on biological systems, particularly at shorter wavelengths. Indeed, many sterilizations and food processing systems use UVC light. On the other hand, small amounts of UVB can be therapeutic for certain skin conditions (e.g., psoriasis; optimum wavelength: ca. 311 nm). In this paper, we report our attempts to use electron/hole pairs bound in excitons as a light source for AlGaIn-based LEDs. Introducing this emission, resulting from an exciton transition, has the potential to provide exciton-related light-emitting devices to replace gas light sources with solid-state light sources.

A large exciton binding energy induces strong excitonic effects in AlGaIn. For example, the optical absorption spectrum exhibits sharp resonance features [4–6], due to dominating direct transitions between the valence and conduction bands at the same K (wave vector) points. [7–9] These strong excitonic effects cause a significant transfer of oscillator strength from the band-to-band transition to the 1s exciton state. [10] The ratio of the oscillator strength of the 1s exciton state to the band-to-band transitions can reach up to 100-fold. [11–13] AlN (exciton binding energy: 80 meV) is not the only material in which excitons have been used as the basis of light-emitting devices. Several other wide-band-gap semiconductors, including diamond (exciton binding energy: ca. 80 meV) and zinc oxide (exciton binding energy: 60 meV), have stable excitons at room temperature. [1] The binding energy for stable excitons must be greater than the equivalent thermal energy at room temperature. The dominant forces binding excitons are the coulombic interactions between the negative and positive charge carriers. There are many factors restricting electrons and holes from forming excitons. For example, the K values of the

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elocities. Thus, excitons can form only at the

bottom of the conduction band and the top of the valence band. If there are too many electrons nearby, however, they will block the coulombic attraction between the electron and the hole required to form the exciton. Generally, exciton emission is observed from relatively pure materials, but not from highly doped materials. [7]

The exciton binding energy is a measure of the stability of the electron/hole pair. An exciton with high binding energy is less likely to be scattered by lattice vibrations to form free charge carriers, or to undergo conversion to free charge carriers as a result of the presence of impurities or defects. Consequently, an exciton having a high binding energy will have a lifetime longer than that of one with a weak binding energy. The higher exciton binding energy also increases the radiative rate and decreases the nonradiative rate. [14] These attributes are important to consider when developing photonic devices. Diamond's exciton, having a binding energy of approximately 80 meV, is stable at room temperature; sharp emission lines at 235 nm (5.27 eV) and 242 nm (5.12 eV) have been attributed to free excitons. [1] Koizumi et al. were the first to report a diamond-based device emitting in the deep-UV; a forward bias of 20 V produced a "strong" UV emission at 235 nm, even though we might rule out diamond as a light emitter because of its indirect band gap. [15] Zinc oxide is another potential exciton light source; its binding energy is approximately 60 meV. Excitation behavior was observed in the UVA band at 390 nm upon irradiating ZnO with 355-nm light from an Nd:YAG solid-state laser, suggesting the possibility of developing a ZnO laser having a low threshold current. Nevertheless, n-type ZnO is relatively easy to prepare, but p-type ZnO is difficult to realize. If a reliable method was available for creating p-type ZnO, ZnO-based devices would presumably have distinct advantages over current commercial GaN LEDs and laser diodes. [16] Taniyasu et al. reported LED wavelengths that were decreased down to 210 nm; they prepared a PIN diode having an electroluminescence wavelength of 210 nm—very close to that of free exciton recombination in undoped AlN in photoluminescence measurements. [17] The AlN exciton has a large binding energy of approximately 80 meV, imparting stability at room temperature. Because of its large activation energy (630 meV), the doping efficiency of Mg-doped p-AlN is very low. [18] Furthermore, no conduction band discontinuity was available to enhance carrier confinement at the AlN homojunction. The output power of the PIN diode reported by Taniyasu et al. was 0.02 μ W at 40 mA. The measured external quantum efficiency (ca. 6–10%) was far from the value of 20% of commercial UVC LEDs. [19]

The biggest problem faced by nitride-based UVC and UVB LEDs is the application of AlN with a high Al content, because the doping activation energy of Mg is very high and the doping efficiency is very low. The thickness of p-AlGaIn must, therefore, be increased to satisfy the requirements of quasi-charge neutrality. However, up to now, the conductivity of the neutral region of AlGaIn DUV still cannot meet the condition of quasi-charge neutrality when injected into the quantum well (excess electrons are injected and then the same number of holes are injected). It will need a longer thermal equilibrium reaction time. And also make the series resistance larger, resulting in bias loss. [20, 21] In addition, because of the effects of light absorption and the series resistance, the thickness of the p-GaN contact layer cannot be greater than 40 nm. Therefore, the mismatch in the degrees of electron/hole injection in the active layer of the quantum well will lead to a lack of quasi-charge neutrality. In addition, the traditional UVB structure

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e quantum well active layer, but it blocks not

only the overflow of electrons but also the injection of holes. The presence of exciton emission can greatly improve the luminous efficiency of an LED device. The exciton binding energies of AlN, diamond, and ZnO are 80, 80, and 60 meV, respectively, but at present it remains difficult to form p–n junctions from these three materials. AlGaN is the most promising material for forming p–n junctions to exploit exciton emissions. Furthermore, the AlN template and sapphire have been the best substrates for UVC and UVB emissions. In addition to the doping activation energy, limitations in heterojunction lattice mismatch epitaxial growth technology have made it more difficult to improve the luminous efficiency in the UVB band than in the UVC band. Therefore, to ensure the exciton emission of nitride-based compounds in the UVB band at room temperature, it will be necessary to shorten the relaxation time of the holes in the neutral region and allow the luminescent layer to reach a state of quasi-charge neutrality. Therefore, by considering the requirements for light absorption and the relaxation time of the holes, we designed an LED containing a 20-nm-thick layer of p-GaN. In addition, before depositing the p-GaN layer, we abandoned the AlGaN EBL and replaced it with a two-fold p-Al_{0.55}GaN/p-Al_{0.4}GaN structure to improve the crystal quality. After depositing the two-fold layer structure, to minimize the two-dimensional electron gas (2DEG) produced by the spontaneous polarization and piezoelectric polarization between materials, which would lead to electron/hole recombination and parasitic luminescence in the neutral region of the p-type structure, we also deposited a 50-nm-thick p-AlGaN–to–p-GaN grading composition layer. Finally, we deposited the 20-nm-thick p-GaN layer to ensure a sufficient number of holes and good ohmic contact. If too many electrons or holes accumulated near the lowest energy of the quantum well, we suspected that a screening effect would break apart the formed exciton pairs. Because exciton emission is generally observed from relatively pure materials, but not from highly doped materials, we did not dope the quantum wells or barriers. Using this design and arrangement, we observed exciton emission behavior from AlGaN-based LEDs. Imagine we were matchmakers trying to combine Mr. Electrons and Miss. Holes to ensure happy marriages, is there any possible approach? With the semiconductor exciton light-emitting device, the answer is affirmative. Because of the Coulomb attraction, the energy of the electron-hole pair as a whole has increased. Additionally, since Electron and Holes are close to each other and with the same velocity, the rate of direct recombination will be faster, improving the efficiency by the exciton emission. In this paper, AlGaN UVB exciton emission by increasing the numbers of Holes. Also, we make some energy sacrifices, photon absorbed, in exchange for the Global green technology.

Experimental Methods

The epitaxial growth of AlGaN-based LEDs was performed using low-pressure metal–organic chemical vapor deposition (LP-MOCVD). Trimethylaluminum, trimethylgallium, silane, and ammonia were used as Al, Ga, Si, and N sources, respectively.

To obtain the UVB LED (sample A), a 2.2-μm-thick AlN buffer layer was first grown on a 2-inch (0001) oriented sapphire substrate. Next, an interlayer consisting of 30 periods of a 2.5-nm AlN/17.5-nm AlGaN superlattice with an equivalent Al composition (0.72) was grown on the AlN buffer layer. A 1.5-μm-thick undoped Al_{0.6}Ga_{0.4}N layer was grown on the superlattice interlayer. Subsequently, a 2-μm-thick Si-

doped $n\text{-Al}_{0.5}\text{Ga}_{0.5}\text{N}$ layer was grown as the n-contact. The active region included four periods of $\text{Al}_{0.35}\text{Ga}_{0.65}\text{N}$ (2 nm)/ $\text{Al}_{0.45}\text{Ga}_{0.55}\text{N}$ (8 nm) multiple quantum wells (MQWs). A two-fold Mg-doped $p\text{-Al}_{0.55}\text{Ga}_{0.45}\text{N}$ (10 nm)/Mg-doped $p\text{-Al}_{0.4}\text{Ga}_{0.6}\text{N}$ (2 nm) structure, replacing the conventional AlGaN EBL, was grown, followed by a 50-nm-thick Mg-doped $p\text{-Al}_{0.3}\text{Ga}_{0.7}\text{N}$ -to- $p\text{-GaN}$ grading layer. Finally, a 20-nm-thick Mg-doped $p\text{-GaN}$ layer was deposited to serve as the p-contact layer and also to modulate the hole relaxation time (Fig. 1).

To obtain the UVC LED (sample B), the AlN buffer layer, 30-period superlattice, and undoped $\text{Al}_{0.6}\text{Ga}_{0.4}\text{N}$ layer were prepared in the same manner as those in sample A. Subsequently, a 2- μm -thick Si-doped $n\text{-Al}_{0.6}\text{Ga}_{0.4}\text{N}$ layer was grown as the n-contact. The active region included four periods of $\text{Al}_{0.45}\text{Ga}_{0.55}\text{N}$ (2 nm)/ $\text{Al}_{0.55}\text{Ga}_{0.45}\text{N}$ (8 nm) MQWs. A two-fold Mg-doped $p\text{-Al}_{0.55}\text{Ga}_{0.45}\text{N}$ (10 nm)/Mg-doped $p\text{-Al}_{0.45}\text{Ga}_{0.55}\text{N}$ (2 nm) structure was then grown, replacing the conventional AlGaN EBL, followed by a 50-nm-thick Mg-doped $p\text{-Al}_{0.3}\text{Ga}_{0.7}\text{N}$ -to- $p\text{-GaN}$ grading layer. Finally, a 20-nm-thick Mg-doped $p\text{-GaN}$ layer was deposited to serve as the p-contact layer and also to modulate the hole relaxation time.

After performing the LP-MOCVD growth processes, the samples were annealed in a N_2 ambient to activate the Mg-dopants. The LED chips were fabricated using standard chip-processing technologies. Mesa structures were defined through inductively coupled plasma etching to expose the $n\text{-Al}_{0.5}\text{Ga}_{0.5}\text{N}$ layer surface. N-contacts of Ti/Al/Ti/Au (100/200/30/100 nm) were deposited through electron-beam evaporation and annealed through rapid thermal annealing at 980°C for 60 s. To form the transparent p-contact, a 50-nm-thick layer of indium tin oxide (ITO) was sputter-deposited and annealed at 600°C for 10 min. The LED chips were completed with the deposition of Ti/Pt/Au (50/30/100 nm) and AuSn (3 μm); the p-contact area was approximately 0.14 mm^2 .

The prepared flip-chips were bonded on an AlN direct plating ceramic lead frame using the eutectic method (AD211 plus, ASM); a covering quartz glass served as an optical lens. The packaged samples were soldered onto an Al metal core printed circuit board. The light output power (LOP), the current–voltage characteristics, and the electroluminescence (EL) spectrum of the samples were measured using an ATA-5000 LED photoelectric measurement system (Everfine) equipped with a 30-cm-diameter integrating sphere. During measurement, the temperature of the heat-sink mounting the packaged sample was controlled between 298 and 368 K at a driving direct current was 40 mA (current density: ca. 28.5 A cm^{-2}). To minimize the effect of self-heating of the chips, the stop interval was set to 3 min during each continuous wave (CW) measurement.

Results And Discussion

Figures 2(a) and 2(b) present the EL band peak positions (measured at room temperature) of the two samples before and after performing burning tests at 40 mA. Sample A exhibited a clear exciton emission, with its peak near 306 nm, and a band-to-band emission at 303 nm. The peak at 306 nm also might be the quantum well composition that causes the spatial potential fluctuations, but we observed

that narrow band full widths at half maximum (FWHM) less than 10nm and the composition fluctuations will result in a larger FWHM, so we prefer this peak was caused by exciton emission. Because the excitons existed in the crystal and not in free space, the binding energy was decreased by the reduced mass and the dielectric constant, where R_y is equal to 13.6 eV and n is an integer greater than or equal to 1.

$$E_x = - \left(\frac{m_r q^4}{2h^2 \epsilon^2} \right) \frac{1}{n^2} = - \frac{(m_r/m_0) R_y}{\epsilon_r^2 n^2}$$

1

After applying Eq. (1), we obtained an exciton bonding energy for $\text{Al}_{0.35}\text{Ga}_{0.65}\text{N}$ of approximately 39 meV. Accordingly, we confirmed that the signal near 306 nm arose from exciton emission. Sample B provided only a Gaussian peak emission at a wavelength of approximately 272 nm. We calculated the Al content in the AlGa_N structure by using the Schrödinger wave equation with a quantized level shift and following Vegard's law; the $\text{Al}_{0.45}\text{Ga}_{0.55}\text{N}$ band-to-band emission was approximately 270 nm. Therefore, we attributed the emission at 272 nm mainly to the band-to-band transition. For both samples, increasing the temperature caused the intensity of the band-to-band emission to decrease and shift to slightly longer wavelength. The relative intensity of the exciton emission in sample A decreased as a result of a thermal energy effect. Sample A provided the exciton emission at temperatures of up to 363 K. Several reports have described parasitic peaks extending from the low-energy side of the main peak for UVC LEDs. [22] These parasitic peaks are presumably associated with recombination occurring through deep level defects and electron overflow and through polarization doping recombination in the p-AlGa_N neutral region. In this present study, we did not observe any obvious parasitic peaks from sample B at any temperature up to 363 K. Therefore, we suggest that defect-related recombination did not occur in sample B under the injection conditions in the temperature range from approximately 298 to 363 K. Accordingly, using the two-fold p- $\text{Al}_{0.55}\text{Ga}_\text{N}$ /p- $\text{Al}_{0.4}\text{Ga}_\text{N}$ structure (to improve the crystal quality), the 50-nm-thick p-AlGa_N-to-p-GaN grading composition layer, and the 20-nm-thick p-GaN layer (to ensure a sufficient number of holes and good ohmic contact) effectively depressed the emission band arising from defects or impurities. Santi et al. reported [23] that the normalized intensity of parasitic peak 3 (ca. 340 nm) from a UVB LED decreased monotonically upon increasing the temperature from 100 to 400 K; they suggested that the peak 3 originated from radiative transitions through deep levels in the quantum barrier next to the EBL, because of its broad shape.

Figures 3(a) and 3(b) reveal the temperature-dependence of the peak wavelengths of samples A and B at 40 mA. The peak wavelengths and full widths at half maximum (FWHMs) of both samples shifted slightly—from 306.0 nm (9.612 nm) to 306.6 nm (11.02 nm) and from 271.8 nm (10.34 nm) to 272.5 nm (11.24 nm), respectively—upon increasing the temperature from 298 to 363 K. Consistent with Varshni's law, the red-shifts were caused by narrowing of the band gap upon increasing the temperature. The

FWHM of the band-to-band emission of the reciprocal of $E_g(T)$, because of

poorer quality resulting from the higher Al content and more alloy scattering layer in the AlGaIn MQW. Furthermore, the density of carriers available for radiative recombination inside in MQWs had decreased.

Sample A was more likely to experience a decrease in the radiation recombination rate and an increase in the non-radiative recombination rate upon increasing the temperature. At temperatures of up to 363 K, the emission of sample A appeared as two Gaussian peaks, the exciton emission and band-to-band emission.

To obtain a deeper understanding of the temperature-dependence of the emissions from the UVB and UVC LEDs, we measured the non-normalized light output power of samples A and B at various temperatures (Fig. 4). Upon increasing the temperature, the light output power from both samples decreased monotonically, but the slope of the line for sample A was much smaller than that for sample B. The exciton emission from sample A was less likely to be scattered by phonons. The photonic device temperature reliability is known to be affected by the carrier confinement factor. A larger discontinuity in the conductive band enhances the thermal stability. Because the values of ΔE_c of samples A and B were nearly identical, the electroluminescence of sample A must have arisen, in part, from the exciton emission. According to the fitting data derived from the temperature-dependence of the light output power, the values of characteristic temperature (T_0) of samples A and B were 328 and 108 K, respectively. Chhajed et al. reported that a larger value of T_0 could decrease the contribution of non-radiative recombination to the total carrier recombination, due to saturation of the non-radiative recombination paths; as a result, the activation energy of the non-radiative recombination centers and the energy required to overcome the confining potentials would both increase. [24]

Ploch et al. [25] investigated the influence of the dislocation density and the barrier height on the temperature-stability of the light output power from a 380-nm LED. The decrease in the temperature-stability was more pronounced in a sample with high TDD (threading dislocation density) and weakly influenced by the barrier height when the current density was less than 50 A cm^{-2} . In this present study, both of our samples had the same structure below the Si-doped n-contact layer, and no indium was present in the MQW active region. Nevertheless, sample A was less temperature-sensitive than sample B in its light output power at a current density of approximately 28.5 A cm^{-2} . Hence, we attribute the improved temperature-stability of sample A to its lower effective TDD and superior carrier confinement in the active region, due to the exciton quantum confine effect. The behavior of sample A can be explained by considering that its exciton emission was less likely to be scattered by lattice vibrations to form free charge carriers, and by its insensitivity to impurities or defects. As mentioned above, limitations in heterojunction lattice mismatch epitaxial growth technology have made it more difficult to improve the luminous efficiency of the UVB band relative to the UVC band. As a result, the TDD in sample A had decreased effectively.

Conclusion

Among the semiconductor materials, two light-emitting materials are considered to have great potential—AlN and ZnO, but each of them has very low p-type doping efficiency and are difficult to prepare. In this paper, we present a new approach toward a nitride-based UVB LED that successfully realizes an exciton emission in the UVB band. Our design suppressed the parasitic emission caused by electron overflow in a typical UVB. In addition, the p-type layer structure and doping profile ensured conditions of quasi-charge neutrality in the UVB quantum well. Despite the content of Al in the luminescent layer of sample A being smaller than that of sample B, sample A was less temperature-sensitive in its the light output power at a current density of approximately 28.5 A cm^{-2} . According to the fitting data derived from the temperature-dependence of the light output power, the characteristic temperatures (T_0) of samples A and B were 328 and 108 K, respectively.

Abbreviations

UVB LED: Ultraviolet-B light-emitting diodes; UVC LED: Ultraviolet-C light-emitting diodes; 2DEG: Two-dimensional electron gas; LP-MOCVD: Low-pressure metal–organic chemical vapor deposition; LOP: Light output power; CW: Continuous wave; FWHM: Full widths at half maximum;

Declarations

Competing interests

The authors declare that they have no competing interests.

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Authors' contributions

M.J. Lai, R.S. Liu designed the experiments. R.M. Lin and C.H. Kuan analyzed data. T.Y. Liu and S.M. Huang discussed the results and contributed to the writing of the manuscript. All authors read and approved the final manuscript.

Available of data and materials

The datasets supporting the conclusions of this article are available in the article

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Figures

Figure 1

Schematic representation of the epitaxial structure of sample A.

(a)

(b)

Figure 2

EL band peak positions, measured at room temperature before and after burning tests, of (a) sample A ($V_f = 5.246$ V @40 mA) and (b) sample B ($V_f = 5.530$ V @40 mA).

(b)

(b)

Figure 3

Temperature-dependence of the EL peak wavelengths of (a) sample A ($V_f = 5.246$ V @40 mA) and (b) sample B ($V_f = 5.530$ V @40 mA).

Figure 4

Light output power from samples A and B at various temperatures at 40 mA.