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# Influence of Quantum-Well Width on the Electroluminescence Properties of AlGaIn Deep Ultraviolet Light-Emitting Diodes at Different Temperatures

Shuxin Tan<sup>1</sup>, Jicai Zhang<sup>2,4\*</sup>, Takashi Egawa<sup>3\*</sup>, Gang Chen<sup>4</sup>, Xiangdong Luo<sup>1</sup>, Ling Sun<sup>1</sup> and Youhua Zhu<sup>1</sup>

## Abstract

The influence of quantum-well (QW) width on electroluminescence properties of AlGaIn deep ultraviolet light-emitting diodes (DUV LEDs) was studied at different temperatures. The maximum external quantum efficiency (EQE) ratios of LED with 3.5 nm QW to that with 2 nm increased from 6.8 at room temperature (RT) to 8.2 at 5 K. However, the ratios for LED with 3.5 nm QW to that with 5 nm QW decreased from 4.8 at RT to 1.6 at 5 K. The different changes of EQE ratios were attributed to the decrease of non-radiative recombination and the increase of volume of the active region. From theoretical analysis, the LED with 2-nm wells had a shallowest barrier for electron overflow due to the quantum-confined effect, whereas the LED with 5-nm wells showed the least overlap of electron and hole due to the large internal field. Therefore, the LED with 3.5 nm QW had the highest maximum EQE at the same temperature. As temperature decreased, the current for maximum EQE decreased for all the LEDs, which was believed to be due to the increase of electron which overflowed out of QWs and the decrease of hole concentration. The results were helpful for understanding the combination of polarization effect and electron overflow in DUV LEDs.

**Keywords:** Deep ultraviolet light-emitting diodes, AlGaIn, External quantum efficiency, Electroluminescence, Low temperature

## Background

AlGaIn-based deep ultraviolet light-emitting diodes (DUV LEDs) can be widely used in the fields of solid-state lighting, medicine, biochemistry, and so on. Therefore, more and more efforts have been devoted to improve the crystal quality of the materials [1–4], the p-type doping techniques, and the optimization of the device structures [5–9]. Miyake et al. demonstrated that the AlN crystal quality can be improved significantly by high-temperature annealing [3]. By increasing the growth temperature, Sun et al. obtained high-quality AlN thick films on sapphire [2]. Recently, Jiang et al. studied the defect evolution in AlN homoepitaxial

growth [1]. Their results contributed to the understanding of the AlN homoepitaxy mechanism and provided the critical techniques for improving the crystal quality. In addition, many methods were proposed to improve the light extraction, such as photonic crystals and nanostructures and surface plasmon [10–12]. In the past decades, a great progress has been obtained for this kind of LED, which was reviewed overall by Li et al. [13]. Nevertheless, the performance of the devices is still far from the practical application due to the low external quantum efficiency. It is well known that group III nitrides have wurtzite structures, in which the large spontaneous and piezoelectric fields will result in the tilted band diagram. These tilted bands had great influence on group-III nitride-based devices, such as LED, LD [14, 15], and UV detectors [16, 17]. Hirayama et al. reported the influence of quantum-well (QW) width on the photoluminescence (PL) properties in AlGaIn-based single-QW DV LEDs [18]. They found that the LEDs

\* Correspondence: jczhang@mail.buct.edu.cn; egawa.takashi@nitech.ac.jp

<sup>2</sup>Department of Physics, College of Science, Beijing University of Chemical Technology, 15 East Road, Beisanhuan, Beijing 100029, China

<sup>3</sup>Research Center for Nano-Device and System, Nagoya Institute of Technology, Gokiso-cho, Showa-ku, Nagoya 466-8555, Japan  
Full list of author information is available at the end of the article

with QW width of 1.5–1.7 nm exhibited a higher luminescence and the PL intensity decreased when the QW width was less than 1.5 nm, which was attributed to an increase of nonradiative recombination on the heterointerfaces. In this work, we fabricated DUV LEDs with different quantum-well (QW) width and studied the influence of the QW width and temperature on the electroluminescence (EL) properties. We found the LEDs with QW width of 3.5 nm exhibited the highest maximum external quantum efficiency (EQE). As the temperature decreased, the current for the maximum EQE decreased for all the LEDs, which was believed to be due to the decrease of hole concentration and the increase of overflowed electron current.

## Methods

The LEDs were grown by metal-organic chemical vapor deposition on (0001)-sapphire substrate using a 1.0- $\mu\text{m}$  AlN buffer layer followed by a 0.5- $\mu\text{m}$ -thick undoped  $\text{Al}_{0.6}\text{Ga}_{0.4}\text{N}$  and a 1.0- $\mu\text{m}$ -thick  $n\text{-Al}_{0.6}\text{Ga}_{0.4}\text{N}$  template. The dislocation density of the template is around  $6 \times 10^9 \text{ cm}^{-2}$  measured by transmission electron microscopy. Then  $\text{Al}_{0.49}\text{Ga}_{0.51}\text{N}/\text{Al}_{0.58}\text{Ga}_{0.42}\text{N}$  multiple QWs (MQWs) were grown as active regions. The thickness of the barriers was 5.0 nm.  $p\text{-Al}_{0.3}\text{Ga}_{0.7}\text{N}$  (25 nm)/ $\text{Al}_{0.6}\text{Ga}_{0.4}\text{N}$  (25 nm) was used as p-type layers. Finally, a 200-nm p-GaN contact layer was deposited. Based on the above structure, three samples, named LEDs A, B, and C, were grown with QW width of 2.0, 3.5, and 5.0 nm, respectively.

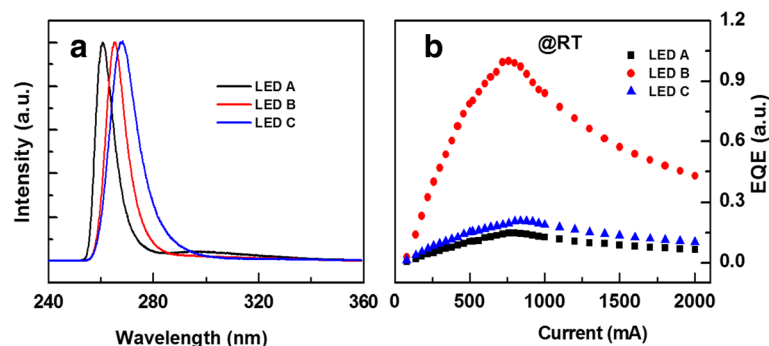
500  $\mu\text{m} \times 500 \mu\text{m}$  square geometry  $p\text{-}n$  junction devices were fabricated using standard lithographic techniques to define the feature and reactive ion etching to expose the  $n\text{-Al}_{0.6}\text{Ga}_{0.4}\text{N}$  Ohmic-contact layer. n-type Ohmic contacts of Ti/Al/Ni/Au (15/80/12/60 nm) were deposited by electron-beam evaporation and annealed with a rapid thermal annealing system at 900  $^{\circ}\text{C}$  for 30 s in nitrogen ambience. For transparent  $p$ -contacts, Ni/Au (6/12 nm) layers were electron-beam deposited and annealed in air ambience at 600  $^{\circ}\text{C}$  for 3 min. The device

was completed with the deposition of Ni/Au (5/60 nm)  $p$  contact. The EL spectrum was measured from 5 K to room temperature (RT) using Jonin Yvon's Symphony UV-enhanced liquid nitrogen-cooled charge-coupled device detector. To avoid the influence of thermal heating effect [19], the pulse injection with 1  $\mu\text{s}$  current pulse at 0.5% was used in the EL measurements.

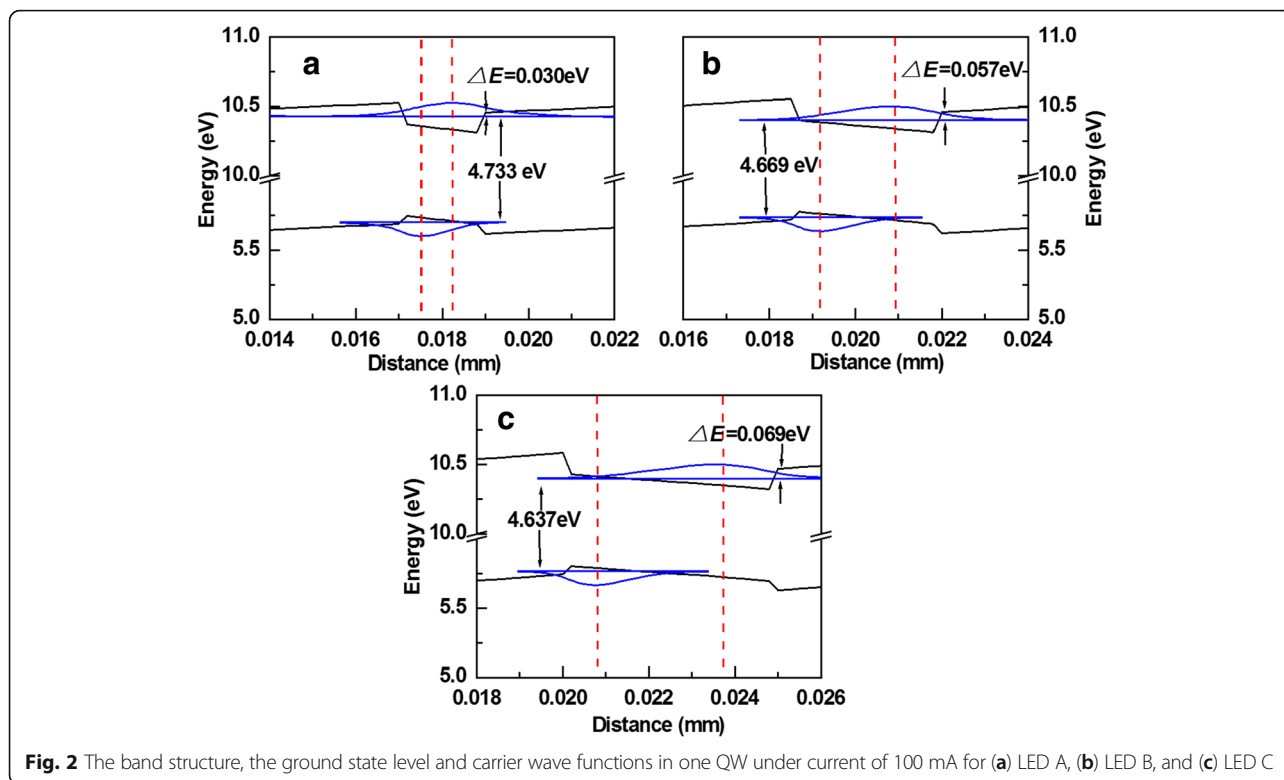
## Results and Discussion

Figure 1a shows the EL spectra measured at room temperature (RT) for LEDs A, B, and C under direct current of 100 mA, in which all the spectra were normalized to the band-to-band emission. The EL peaks for LEDs A, B, and C were around 261, 265, and 268 nm, respectively. Obviously, the EL peak showed a redshift as the QW width increased. In addition, it should be noticed that a weak parasitic peak around 304 nm existed in the EL spectrum in LED A, which was clarified to be related with electron overflow [20]. Figure 1b shows the relative EQE as a function of pulse current for all the LEDs. All the values were normalized to the maximum EQE of LED B. The maximum EQE of LED B was about 6.8 and 4.8 times than those of LEDs A and C, respectively.

In order to understand the reason, APSYS was used to simulate the energy levels and wave functions of carriers. Figure 2a–c shows the band structures, the ground state level, and carrier wave functions in one QW under current of 100 mA for LEDs A, B, and C, respectively. Due to the large internal field induced by the polarization effect and the applied forward bias, the band structure of QW showed an inclined shape and the spatial overlapping of wave functions of electrons and holes became less as the QW width increased due to the quantum-confined Stark effect (QCSE). The energy gap of the ground states for LEDs A, B, and C were 4.733, 4.669, and 4.637 eV, respectively, which coincided well with the emission wavelength as shown in Fig. 1a. In addition, it should be noticed that the confined ability of carriers by the QWs decreased as the QW width



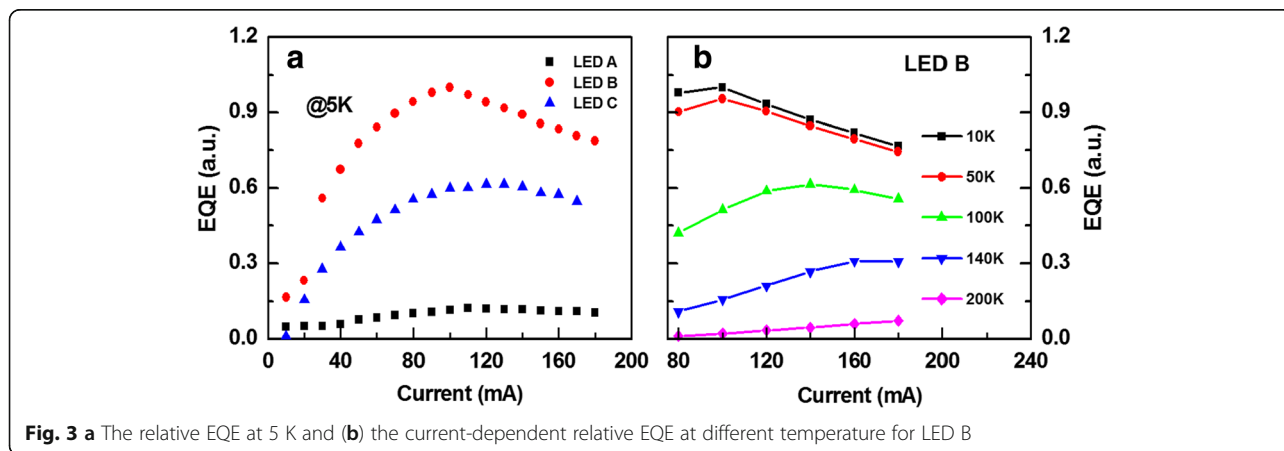
**Fig. 1** a The RT EL spectra for LEDs A, B, and C under direct current of 100 mA. All the spectra were normalized to the band-to-band emission. b The relative EQE as a function of pulse current



decreased. The quantum-confined effect resulted in the increase of the ground state level as the QW width decreased. The values of barrier height were 0.030, 0.057, and 0.069 eV for LEDs A, B, and C, respectively. Therefore, the EQE of LED A was less than that of LED B due to the electron current overflow, which could be confirmed by the obvious parasitic peak shown in Fig. 1a. Though LED C had the highest barrier for electron overflow in all the devices, its EQE was still less than that of LED B due to the QCSE.

The EQE at low temperature was measured to evaluate the device performance. Figure 3a shows the relative EQE measured at 5 K. All the values were normalized to

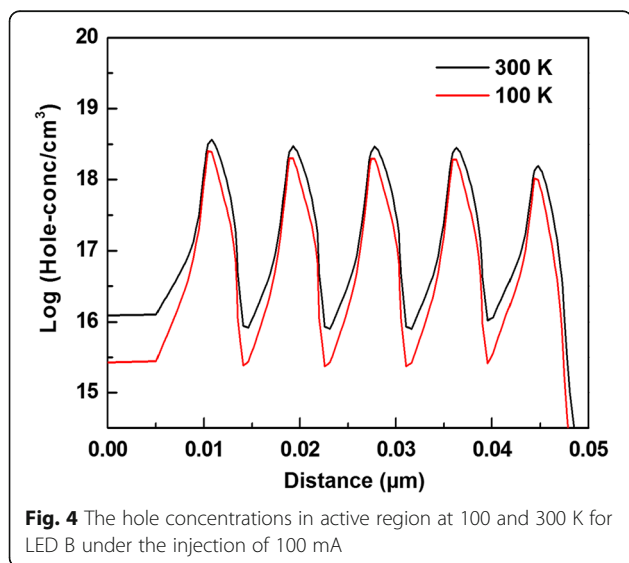
the maximum EQE of LED B. Obviously, the injection current for the maximum EQE decreased significantly compared to those at RT for all the devices. The maximum EQE of LED B was about 8.2 and 1.6 times than those of LEDs A and C, respectively. The current-dependent EQE were measured at different temperature. Figure 3b shows the current-dependent relative EQE at different temperature for LED B. All the values were normalized to the maximum EQE at 10 K. It can be seen that the current for the maximum EQE decreased as the temperature decreased. The same phenomenon was found for all three LEDs. It was well known that in bulk materials the hole concentration



would decrease rapidly with decreasing temperature due to the high ionization energy of Mg in p-AlGa<sub>N</sub>. In our structure, it was demonstrated that the hole concentration also decreased as the temperature decreased [21]. We also simulated the hole distribution at different temperature. Figure 4 shows the hole concentrations in active region at 100 and 300 K for LED B under the injection of 100 mA. Obviously, the hole concentration decreased as the temperature decreased. In addition, the electron current overflowed out of QWs can be expressed as [22].

$$J_{\text{overflow}} = D \left( \frac{\Delta E}{kT} \right)^3 qBl$$

where  $D$  is a constant,  $\Delta E$  is the difference of Fermi level and band edge of quantum wells,  $K$  is the Boltzmann constant,  $T$  is the temperature,  $q$  is the electron charge,  $B$  is the bimolecular radiative recombination coefficient, and  $l$  is the thickness of MQWs. For a certain LED, the contribution of variation of  $\Delta E$  to  $J_{\text{overflow}}$  could be neglected compared to that of  $T$  as the temperature decreased. Therefore, the  $J_{\text{overflow}}$  increased significantly at 5 K compared to that of RT, which was believed to be the main reason for the decrease of injection current at which the maximum EQE reached. The  $J_{\text{overflow}}$  decreased as the temperature increased, resulting in the increase of injection current for the maximum EQE, as shown in Fig. 3b. At low temperature, the internal efficiency would increase due to the freeze-out of nonradiative centers, such as the dislocations, which was beneficial to LED C with the largest volume of active region. This was the most possible reason why the EQE ratio of LED B to LED C decreased at 5 K compared to that at RT. Similarly, the EQE ratio of LED B to LED A increased at 5 K compared to that at RT.



**Fig. 4** The hole concentrations in active region at 100 and 300 K for LED B under the injection of 100 mA

## Conclusions

We studied the influence of QW width on EL properties of AlGa<sub>N</sub> DUV LEDs at different temperatures. The EL spectra showed a redshift as the QW width increased. The maximum EQE for LED with QW width of 3.5 nm was about 6.8 and 4.8 times than those of 2 and 5 nm at RT, respectively. However, these values changed to be 8.2 and 1.6 at 5 K, respectively. The different changes of maximum EQE ratios were attributed the decrease of non-radiative recombination and the increase of volume of the active region. From the theoretical analysis, the LED with 2-nm wells showed a shallowest barrier for electron overflow due to the quantum-confined effect, whereas the LED with 5-nm wells showed a least overlap of electron and hole due to the large internal field. Therefore, the LED with 3.5 nm QWs showed the highest maximum EQE. As the temperature decreased, the current for the maximum EQE decreased for all LEDs, which was believed to be due to the increase of electron overflowed out of QWs and the decrease of hole concentration. The maximum EQE for LED with QW width of 3.5 nm was about 8.2 and 1.6 times of those of 2 and 5 nm at 5 K, respectively, which was believed to be due to the decrease of non-radiative recombination centers and the increase of volume of active region.

## Abbreviations

DUV LEDs: Deep ultraviolet light-emitting diodes; EL: Electroluminescence; EQE: External quantum efficiency; MQW: Multiple quantum well; PL: Photoluminescence; QCSE: Quantum-confined Stark effect; QW: Quantum well; RT: Room temperature

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## Availability of Data and Materials

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

## Authors' Contributions

ST did the simulations and wrote and revised the manuscript. JCZ and TE supervised the research work and revised the manuscript. GC repeated the simulations. XDL and LS contributed to discussion of the results and manuscript revision. YHZ contributed to the device process. All authors read and approved the final manuscript.

## Competing Interests

The authors declare that they have no competing interests.

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**Author details**

<sup>1</sup>School of Electronics and Information, Nantong University, 9 Seyuan Road, Nantong 226019, China. <sup>2</sup>Department of Physics, College of Science, Beijing University of Chemical Technology, 15 East Road, Beisanhuan, Beijing 100029, China. <sup>3</sup>Research Center for Nano-Device and System, Nagoya Institute of Technology, Gokiso-cho, Showa-ku, Nagoya 466-8555, Japan. <sup>4</sup>Suzhou Institute of Nano-Tech and Nano-Bionics, CAS, 398 Ruoshui Road, SEID, SIP, Suzhou 215123, China.

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**References**

- Jiang K, Sun X, Ben J, Jia Y, Liu H, Wang Y, Wu Y, Kai C, Li D (2018) The defect evolution in homoepitaxial AlN layers grown by high-temperature metal-organic chemical vapor deposition. *CrystEngComm* 20:2720–2728
- Sun M, Zhang J, Huang J, Li X, Wang L, Liu X, Wang J, Xu K (2016) Influence of thickness on strain state and surface morphology of AlN grown by HVPE. *J Semicond* 37(12):123001
- Miyake H, Lin C-H, Tokoro K, Hiramatsu K (2016) Preparation of high-quality AlN on sapphire by high-temperature face-to-face annealing. *J Cryst Growth* 456:155–159
- Kim H, Yoon HJ, Choi BJ (2018) Thickness dependence on interfacial and electrical properties in atomic layer deposited AlN on c-plane GaN. *Nanoscale Res Lett* 13:232
- Hirayama H, Maeda N, Fujikawa S, Toyoda S, Kamata N (2014) Recent progress and future prospects of AlGaIn-based high-efficiency deep-ultraviolet light-emitting diodes. *Jap J Appl Phys* 53:100209
- Kinoshita T, Obata T, Yanagi H, Inoue S (2013) High p-type conduction in high-Al content Mg-doped AlGaIn. *Appl Phys Lett* 102:012105
- Li S, Ware M, Wu J, Minor P, Wang Z, Wu Z, Jiang Y, Salamo GJ (2012) Polarization induced pn-junction without dopant in graded AlGaIn coherently strained on GaN. *Appl Phys Lett* 101:122103
- S, Zhang T, Wu J, Yang Y, Wang Z, Wu Z, Chen Z, and Jiang Y Polarization induced hole doping in graded Al<sub>x</sub>Ga<sub>1-x</sub>N (x = 0.7 ~1) layer grown by molecular beam epitaxy *Appl Phys Lett* 2013; 102: 062108
- Zhang ZH, Huang Chen SW, Chu C, Tian K, Fang M, Zhang Y, Bi W, Kuo HC (2018) Nearly efficiency-droop-free AlGaIn-based ultraviolet light-emitting diodes with a specifically designed superlattice p-type electron blocking layer for high mg doping efficiency. *Nanoscale Res Lett* 13:122
- Zhang P, Li S, Liu C, Wei X, Wu Z, Jiang Y, Chen Z (2014) Near-infrared optical absorption enhanced in black silicon via ag nanoparticle-induced localized surface plasmon. *Nanoscale Res Lett* 9:519
- Li D, Sun X, Jia Y, Stockman MI, Paudel HP, Song H (2017) Direct observation of localized surface plasmon field enhancement by Kelvin probe force microscopy. *Light* 6:e17038
- Inoue SI, Naoki T, Kinoshita T, Obata T, Yanagi H (2015) Light extraction enhancement of 265nm deep-ultraviolet light-emitting diodes with over 90 mW output power via an AlN hybrid nanostructure. *Appl Phys Lett* 106: 131104
- Li D, Jiang K, Sun X, Guo C (2018) AlGaIn photonics: recent advances in materials and ultraviolet devices. *Adv Opt Photon* 10:43–110
- Piprek J, Moe CG, Keller SL, Nakamura S, DenBaars SP (2004) Internal efficiency analysis of 280-nm light emitting diodes. *Proc SPIE* 5594:177
- Gotoh H, Tawara T, Kobayashi Y, Kobayashi N, Saitoh T (2003) Piezoelectric effects on photoluminescence properties in 10-nm-thick InGaIn quantum wells. *Appl Phys Lett* 83:4791
- Bao G, Li D, Sun X, Jiang M, Li Z, Song H, Jiang H, Chen Y, Miao G, Zhang Z (2014) Enhanced spectral response of an AlGaIn-based solar-blind ultraviolet photodetector with Al nanoparticles. *Opt Express* 22:24286
- Li D, Sun X, Song H, Li Z, Chen Y, Jiang H, Miao G (2012) Realization of a high-performance GaIn UV detector by nanoplasmonic enhancement. *Adv Mater* 24:845–849
- Hirayama H, Enomoto Y, Kinoshita A, Hirata A, Aoyagi Y (2002) Efficient 230–280 nm emission from high-Al-content AlGaIn-based multi quantum wells. *Appl Phys Lett* 80:37
- Zhang JC, Zhu YH, Egawa T, Sumiya S, Miyoshi M, Tanaka M (2008) Influence of pulse width on electroluminescence and junction temperature of AlInGaIn deep ultraviolet light-emitting diodes. *Appl Phys Lett* 92:191917
- Zhang JC, Zhu YH, Egawa T, Sumiya S, Miyoshi M, Tanaka M (2008) Suppression of the subband parasitic peak by 1 nm i-AlN interlayer in AlGaIn deep ultraviolet light-emitting diodes. *Appl Phys Lett* 93:131117
- Zhang JC, Sakai Y, Egawa T (2010) Low-temperature electroluminescence quenching of AlGaIn deep ultraviolet light-emitting diodes. *Appl Phys Lett* 96:013503
- Schubert EF (2006) *Light emitting diodes*. Cambridge University Press, Cambridge

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