Optical degradation of InGaN/AlGaN light-emitting diode on sapphire substrate grown by metalorganic chemical vapor deposition

T. Egawa
Research Center for Micro-Structure Devices, Nagoya Institute of Technology, Gokiso-cho, Showa-ku, Nagoya 466, Japan

H. Ishikawa
Department of Electrical and Computer Engineering, Nagoya Institute of Technology, Gokiso-cho, Showa-ku, Nagoya 466, Japan

T. Jimbo
Research Center for Micro-Structure Devices, Nagoya Institute of Technology, Gokiso-cho, Showa-ku, Nagoya 466, Japan

M. Umeno
Research Center for Micro-Structure Devices, Department of Electrical and Computer Engineering, Nagoya Institute of Technology, Gokiso-cho, Showa-ku, Nagoya 466, Japan

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We report an optical degradation of an InGaN/AlGaN double-heterostructure light-emitting diode (LED) on a sapphire substrate grown by metalorganic chemical vapor deposition. Electroluminescence, electron-beam induced current, and cathodoluminescence observations have shown that the degraded InGaN/AlGaN LED exhibits formation and propagation of dark spots and a crescent-shaped dark patch, which act as nonradiative recombination centers. The values of degradation rate under injected current density of 0.1 kA/cm^2 were determined to be 1.1×10^{-3}, 1.9×10^{-3}, and $3.9\times10^{-3}\,h^{-1}$ at ambient temperatures of 30, 50, and 80 °C, respectively. The activation energy of degradation was also determined to be 0.23 eV. © *1996 American Institute of Physics.* [S0003-6951(96)04332-X]

Wide-band-gap III–V nitrides and ZnSe-based II–VI compound semiconductors have attracted much attention because their large direct band gap at room temperature is appropriate for short wavelength light-emitting diodes (LEDs) and laser diodes. Stimulated emission has been observed from pulsed current injected GaN-based multiquantum well structure[1] and double-heterostructure (DH) LED with Al reflectors.[2] In particular, recent studies on III–V nitrides have been focused on room-temperature continuous-wave operation of the laser diode since pulsed current operation has been achieved by Nakamura *et al.*[3]

It is widely recognized that high density of dislocations, which act as nonradiative recombination centers, are introduced in epitaxial layers grown by use of the heteroepitaxial growth technique. Dislocations migrate during device operation under high injected current density and ambient temperature, and result in limited state operation of optical devices. For example, GaAs-based laser diodes on the Si substrate, which involve differences of lattice constants and thermal expansion coefficients between GaAs and Si materials, suffer from rapid degradations due to high dislocation density ($>10^6$ cm^{-2}) and large tensile stress ($\sim10^9$ dyn/cm^2) in an active region. We have shown that rapid degradations in AlGaAs/GaAs single quantum well laser diodes on Si substrates are caused by formation of dark-line defects (DLDs) and degraded current–voltage (I–V) characteristic during higher injected current density.[4] Guha *et al.* reported that major degradation in II–VI blue-green light-emitting device occurred due to microstructural changes such as the formation of dark spots, <100> DDLs and dark patches, acting as nonradiative recombination centers.[5] Hua *et al.* also reported that formation of dislocation networks in quantum well region by climb motion of dislocations degraded the characteristics of the II–VI blue-green laser diode during the current injection, which was suggestive of dislocation network formation in degraded AlGaAs/GaAs DH laser diodes.[6] Thus, the degradation of optical characteristic is caused by the dislocations in the epitaxial layer. On the other hand, Lester *et al.* reported that the high density of dislocations ($2-10\times10^{10}$ cm^{-2}) in GaN-based LED on sapphire substrates do not act as efficient minority carrier recombination sites in comparison to other III–V materials.[7] We have confirmed that the degraded characteristics of InGaN/AlGaN LED arise from the deterioration

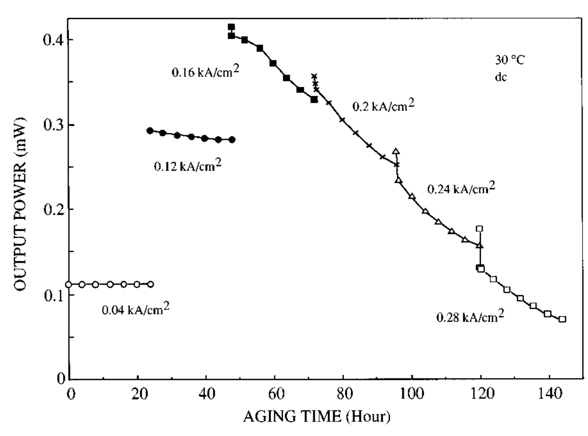

FIG. 1. Variation of output power from InGaN/AlGaN LED as a function of aging time under various injected current densities. Each aging test was performed for 24 h at 30.°C.

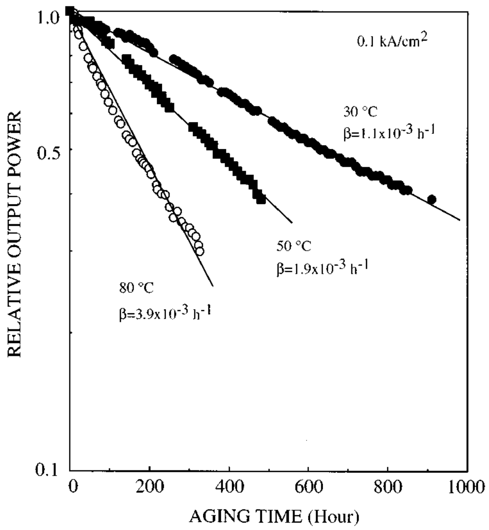

FIG. 2. Variation of relative output power from InGaN/AlGaN LED as a function of aging time at ambient temperatures of 30, 50, and 80 °C. The injected current density was 0.1 kA/cm^2.

of Ohmic electrode and the generation of dark-spot defects (DSDs). In this letter, we focus on an optical degradation of InGaN/AlGaN LED on sapphire substrate under high dc density and high ambient temperature.

The sample in this study was grown on a sapphire substrate with (0001) orientation (c face) by metalorganic chemical vapor deposition (MOCVD) at atmospheric pressure using a modified two step growth technique. After the substrate was heated at 1050 °C in a hydrogen ambient, the InGaN/AlGaN DH was grown. The structure consists of the following growth sequence: a 25-nm-thick GaN buffer layer at 500 °C, a 4-μm-thick n-GaN layer at 1020 °C, a 150-nm-thick n-$Al_{0.15}Ga_{0.85}N$ layer at 1020 °C, a 50-nm-thick $In_{0.06}Ga_{0.9}N$ layer at 780 °C, a 150-nm-thick$p$-$Al_{0.15}Ga_{0.85}N$ layer at 1020 °C, and a 350-nm-thick p-type GaN cap layer at 1020 °C. After the growth, the sample was partially etched until the n-GaN layer was exposed using reactive ion etching. The Ohmic electrodes of Ni/Au and Ti/Al were formed by vacuum evaporation on the p- and n-GaN layers, respectively.[2]

Aging tests were performed under various dc densities and ambient temperatures. Studies of optical degradation were carried out by electroluminescence (EL), electron-beam induced current (EBIC) and cathodoluminescence (CL) methods. EL imaging, to study the formation and propagation of nonradiative recombination centers, was carried out by passing the light exiting from the top surface through the thin Ni pad of the InGaN/AlGaN LED. The degraded samples were also studied by EBIC and CL measurements at accelerating voltage of 20 kV.

The InGaN/AlGaN LED exhibited an optical output power of 0.17 mW, external quantum efficiency of 0.2%, and the peak emitting spectrum at 437 nm with full width at half-maximum of 63 nm at 30 mA (0.06 kA/cm^2). Other characteristics described in previously reported results.[2] Figure 1 shows the variation of output power as a function of aging time under various injected current densities. Each aging test was performed for 24 h under constant current densities from 0.05 to 0.28 kA/cm^2 at 30 °C. Although a gradual decrease was observed in the output power for the injected current density of 0.12 kA/cm^2, stable operation was obtained for lower injected current densities. However, the output power from the sample tested under higher injected current densities decreased rapidly in a few minutes. For an injected current density of 0.28 kA/cm^2, the output power

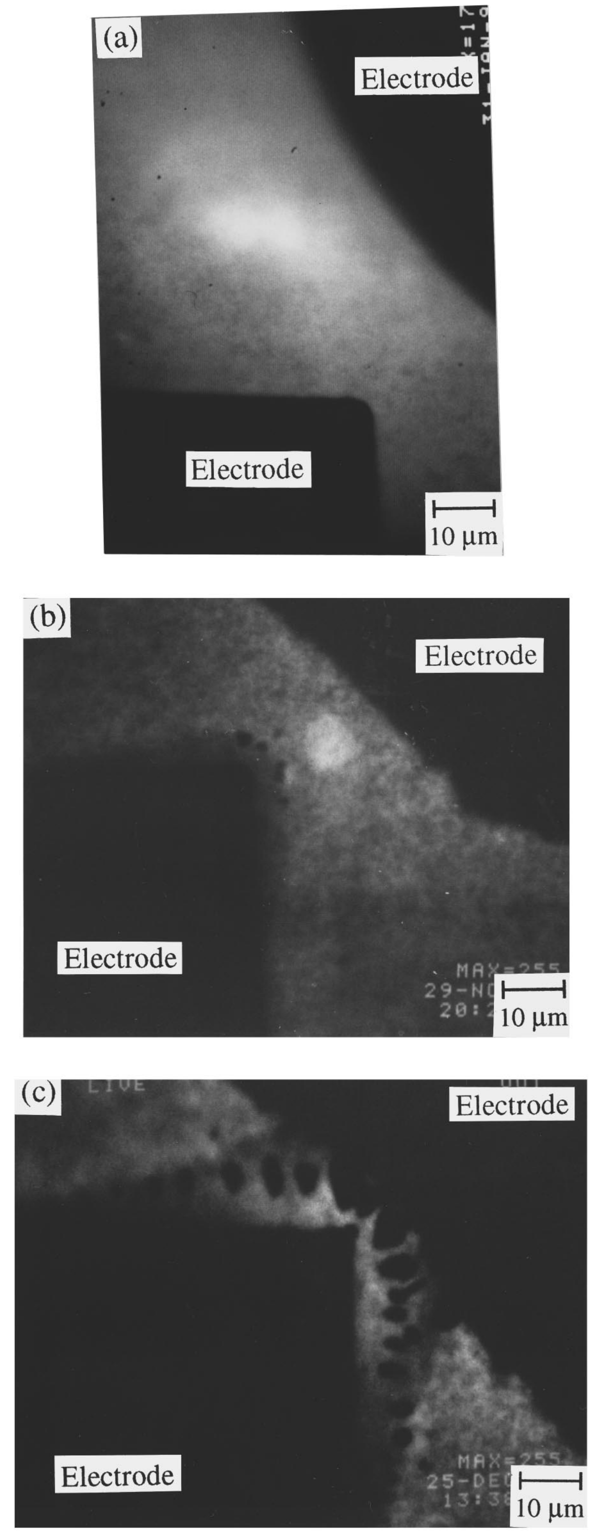

FIG. 3. EL images of progressive statges off degradation for InGaN/AlGaN LED during the aging test under 0.4 kA/cm^2 at 30 °C, (a), (b), and (c) correspond to initial stage and aging of 67 and 310 h, respekctively.

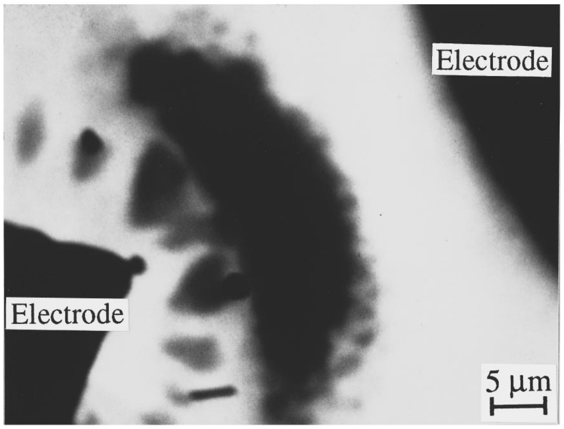

FIG. 4. EBIC image of degraded InGaN/AlGaN LED shown in Fig. 3(c).

initially decreased rapidly, from 0.18 to 0.13 mW in 1 min, and then decreased to 0.07 mW. The light output power-injected current ($L-I$) characteristics were also measured after each aging test was finished. The output power and the external quantum efficiency measured at 30 mA (0.06 kA/cm^2) were 0.17 mW and 0.2% at initial stage, and 0.07 mW and 0.08% after aging at 0.28 kA/cm^2 for 24 h. To investigate the degradation, accelerated aging tests at ambient temperatures of 30, 50, and 80 °C were carried out under the injected current density of 0.1 kA/cm^2. The half-density lifetimes obtained from Fig. 2 were 656.7, 365.7 and 170 h at the ambient temperatures of 30, 50, and 80 °C, respectively. The output power of P can be expressed by[8]

$$P = P_0 \cdot \exp(-\beta t)$$

where P_0, β, and t are the initial output power, the degradation rate, and operating time, respectively. The degradation rate depends on the device temperature, and is given by[8]

$$\beta = \beta_0 \cdot \exp(-E_a/kT)$$

where β_0, E_x, T, and k are a constant, the activation energy of degradation, the device temperature, and Boltzmann's constant, respectively. The values of β were estimated to be 1.1×10^{-3}, 1.9×10^{-3}, and 3.9×10^{-3} h^{-1} at 30, 50, and 80 °C, respectively. The activation energy, E_a, and the value of β_0 were determined to be 0.23 eV and 7 h^{-1}, which were much smaller than the values of 1.0 eV and 1.84×10^7 h^{-1} for InGaAsP LED and 0.57 eV and 93 h^{-1} for AlGaAs LED.[8,9] The temperature rise due to the operating current was not taken into account because of relatively lower injected current density. Thus, the output power decreases during the aging test under higher injected current density and ambient temperature.

In order to study the optical degradation process, EL and EBIC observations were carried out on the InGaN/AlGaN LED. Figures 3(a)–3(c) show the EL images of the progressive stages of degradation during the aging test under 0.4 kA/cm^2 at 30 °C. Figure 3(a) shows the faint dark spots at initial stage, which indicate the pre-existing defects in the structure since they act as nonradiative recombination centers. At the first stage of degradation shown in Fig. 3(b), the faint dark spots become darker and a dark region appears in the vicinity of the corner of the left electrode. In the final stage of degradation shown in Fig. 3(c), the dark spots enlarge individually and the dark region also enlarges. The reason why the dark spots and region were observed in the vicinity of the corner of the left electrode is that the injected current was concentrated at that location. The growth rate of dark spot at 0.4 kA/cm^2 was estimated to be 0.02–0.04 μm/h. We also carried out EBIC and CL measurements on the degraded InGaN/AlGaN LED. Figure 4 shows the EBIC image of the degraded LED observed by the EL image shown in Fig. 3(c). The EBIC image showed that dark spots and a crescent-shaped dark patch were observed, which indicate nonradiative recombination centers in the active region.[5] We also confirmed the dark spots and the crescent-shaped dark patch by the CL method.

In summary, we observed the formation and propagation of the dark spots and a crescent-shaped dark patch in the degraded InGaN/AlGaN LED on the sapphire substrate grown by MOCVD technique. The decrease in the output power under high injected current density and ambient temperature is thought to be caused by the formation of dark regions, which act as nonradiative recombination centers.

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