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Evolution of crystal imperfections during current-stress ageing tests of green InGaN light-emitting diodes

Yue Lin1, Zhangbao Peng1, Lihong Zhu1*, Wei Yan1, Tien-mo Shih2, Tingzhu Wu1, Yijun Lu1*, Yulin Gao1, Zhong Chen1, Ziquan Guo1, and Zhuguang Liu5

1Department of Electronic Science, Fujian Engineering Research Center for Solid-State Lighting, Collaborative Innovation Center for Optoelectronic Semiconductors and Efficient Devices, Xiamen University, Xiamen 361005, China
2Department of Physics, Xiamen University, Xiamen 361005, China
3Fujian Institute of Research on the Structure of Matter, Chinese Academy of Sciences, Fuzhou 350002, China
4E-mail: lhzhu@xmu.edu.cn; yjlu@xmu.edu.cn

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We perform ageing tests under high current on several green InGaN light-emitting diodes and compare the luminous homogeneities of chip surfaces, shapes of external quantum efficiency (EQE) curves, and electroluminescence spectra during different ageing stages. By curve fittings to the EQE curves, with the ABC and two-level models, we discover that a high injection current density can modify the defect configuration in quantum wells even at room temperature, as high-temperature annealing can. For In-rich devices, the removal of localization centers is another origin of luminous intensity decay in addition to the formation of point defects. © 2016 The Japan Society of Applied Physics

Recently, much research work has been focused on the mechanism of the “green gap” and especially on mechanisms of internal quantum efficiency (IQE) droop.1-3 It is widely believed that the high density of structural defects induced by the high indium composition is the primary reason behind these two drawbacks.3 On the other hand, there exists increasing evidence that indium atoms tend to segregate and form clusters, which may increase the local luminous intensity in vicinities.4 Indium fluctuation is considered to be among the main courses of a strong carrier localization effect.5 However, indium atoms would diffuse randomly under high-temperature annealing (>800 °C).1,7 Current-stress or thermal-current-coast tests have been utilized as a tool for investigating the microstructure of InGaN devices.8-14 Interestingly, on the basis of the results of the short-term dc-stress ageing of blue LEDs, Meneghini’s group proposed a hypothesis that the current injected into the active region with a density of 32 A/cm2 was capable of modifying defects at room temperature (RT), leading to the decrease in EQE at low currents.8 In their recent work, they further mention that, in blue InGaN LEDs, the formation of point defects (PDs) is the main reason for the observed luminous intensity.12,13 In our previous work, through monitoring the EQE versus forward current curves (EQE curves) and the current–voltage (J–V) curves, we suggested that such defect modification occurred during current-stressed ageing tests.1,4,15 In this study, by subjecting InGaN/GaN MQW green LEDs to a short-term high-current-stressed ageing test and analyzing the changes in EQE curves and electroluminescence spectra (ELs), we demonstrate that the high current injection greatly affects the microstructure of crystal imperfections in MQWs. By comparing the experimental results from ELs and EQE droop curves, we confirm that, for In-rich devices, besides the increasing PD density, the removal of localization centers plays another key role in giving rise to severe luminous decay during current-stress ageing. Therefore, it is imperative for LED manufacturers to take into account the long-term evolution of crystal imperfections in stages of designing and fabricating devices with high indium composition.

Five InGaN/GaN MQW green LEDs were employed for the ageing test at RT and $I_f = 850$ mA ($J_f = 85$ A/cm2) on a ZWL-190 LED ageing system (Hangzhou Zhongwei Photovoltaic). The device with a vertical structure was grown on a c-axis sapphire substrate by metal organic chemical vapor deposition. In each device, there are 10 pairs of InGaN/GaN multiple quantum wells (MQWs) serving as the active region, sandwiched between n- and p-GaN layers. The structures of all the samples are identical, similarly to those indicated in Ref. 14, but with higher indium compositions. Measurements of EQE curves and forward voltages were carried out before ageing (0h) and at 25, 50, 75, 120, 192, and 336h during ageing. Data were recorded using Spectro 320 (Instrument Systems) in a series of $I_f$, ranging from 0.1 to 850 mA ($J_f = 0.01$–85 A/cm2), provided by Keithley 2400.

We took microscopic pictures of the same sample subjected to $I_f = 90$ μA ($J_f = 0.009$ A/cm2) before and after 50h ageing. As shown in Figs. 1(a) and 1(b), ageing significantly dims the chip. For fresh chips, there exist numerous bright spots lying around the cathode or scattering in areas remote from the cathode; for aged chips, except those around the cathode, most of the bright spots have diminished, leaving a homogeneous luminous surface. Jeong et al. have observed a similar luminous inhomogeneity, which they deem related to the localization effect caused by In-rich areas.59

As shown in Fig. 1(c), the normalized ELs, captured before and after 25h ageing both at 0.1 mA ($J_f = 0.01$ A/cm2), exhibit a conspicuous blue shift by ~5 meV, accompanied by a decrease in linewidth by ~20 meV measured at full width at half maximum. After a careful inspection, it can be considered that such shrinking only occurs on the low-energy side. The band tail that is typically associated with a band-structure associated with an individual defect to several µm.17 Likewise, in this case, the neighboring carriers around an In-rich area within the diffusion length will be bonded to the
energy trap and recombine radiatively near the band edge, increasing the luminous intensity of the area nearby, and leading to a bright spot sufficiently large to be observed. During aging, the crystal structures of In-rich areas will be somehow altered under the constant impact of high Jf stress. Although experimental results in this work are unable to reveal such a modification in detail, they clearly show its aftermath—potential fluctuations are mitigated as well as the localization effect, whereas the homogeneities of the energy band are enhanced. These will result in both the blue shift and linewidth shrinking in ELs at the same time, as already shown in Fig. 1(c).

As a powerful tool for evaluating defect dynamics,\(^\text{14,15}\) EQE curves are measured and illustrated in Fig. 2(a). This figure shows that EQEs decrease markedly at the initial 25 h, especially for low currents, and that the EQE droop onset shifts towards higher currents. The 25 h decay rate also changes with current, as it first increases from 0.01 A/cm\(^2\), peaking at \(\sim 0.1\) A/cm\(^2\) and exceeds 40\%, followed by a monotonic droop with current. We attribute the increase at low currents to the diminishing localization effect. Before ageing, the localization effect occurs at \(J_f\) below 0.1 A/cm\(^2\), which means that at such a \(J_f\), carriers tend to drift or diffuse into the localization centers induced by In-rich areas where they take part in the band-edge emission efficiently.\(^\text{15}\) During ageing, as the localization effect diminishes, carriers are no longer localized when \(J_f\) reaches the same level, but are instead captured by PDs, indicating that the severest decrease in EQE occurs at \(J_f\), which is associated with the largest localization effect. The EQE curves are normalized and shown in a linear scale in Fig. 2(b) to expose the falling segment, the slope of which becomes gentler rather than steeper, as ageing proceeds.

To extract defect information from EQE curves, we employ the ABC model. As introduced by Schubert’s group, this model has been well used in the evaluation of EQE curves.\(^\text{18,19}\) Utilizing the ABC model to evaluate EQE curves during ageing has been reported in detail.\(^\text{12}\) The equation of the ABC model is shown as

\[
\eta_{\text{EQE}} = \frac{\eta_{\text{ext}} \eta_{\text{inj}} B n^2}{A n^2 + B n^2 + C n^2}.
\]

In Eq. (1), \(n\) denotes the carrier concentration, which is considered to be proportional to the square root of light output power \((\sqrt{\text{LOP}})\).\(^\text{19}\) \(\eta_{\text{ext}}\) and \(\eta_{\text{inj}}\) denote the light extraction and carrier injection rates, respectively. The parameters \(A\), \(B\), and \(C\) are the coefficients of SRH nonradiative recombination, band-edge recombination, and Auger recombination, respectively. The absolute values of these three parameters cannot be obtained by curve fitting at the same time. In this work, we perform the curve fitting of normalized EQE curves with respect to \(\sqrt{\text{LOP}}\) obtained when measuring EQE curves, and compare the relative changes in \(A\) and \(C\) with ageing, by considering that \(B\) remains constant during ageing. A curve fitting via the ABC model is illustrated in Fig. 2(c) using a green line; it can be observed that the fitting accuracy of the rising section is higher than that of the falling section. Plots of \(A\) vs ageing time are shown in Fig. 2(d), where \(A\) increases with ageing, indicating an increase in SRH nonradiative recombination intensity, as we have expected. \(A\) increases rapidly at the first 100 h and then slowly. This observation is different from that reported in Ref. 12, that is, an almost linear increase with ageing time. This may be due to the fact that they stress those devices at 100 mA, a milder condition than ours (850 mA). Compared with \(A\), \(C\) nonetheless exhibits an irregular shift (thus not shown), which we ascribe to the low fitting accuracy at a high current (corresponding to a high carrier concentration).

As an alternative way of evaluating only the rising segment, we introduce the two-level model, the equation of which is written as\(^\text{14,15}\)

\[
\eta_{\text{EQE}} = \frac{C \left[ 1 - (\alpha + \beta)/J_f \right]^{1/2}}{2 + \sqrt{4\alpha/J_f + (1 - (\alpha + \beta)/J_f)^2}}.
\]

In the two-level model, the competition between band-edge recombination and SRH recombination is taken into account. In other words, once generated, carriers either recombine radiatively at the band edge or are trapped into nonradiative SRH defects, which are usually PDs. In Eq. (1), \(C\) is a constant, depending on the light extraction rate of the device; \(\alpha = W_i/W_i/\gamma_i\) and \(\beta = N_i W_i\), where \(W_i\) and \(W_f\) denote the interband radiative and nonradiative recombination rates via PDs, respectively, \(\gamma_i\) the defect capture rate of an individual PD, and \(N_i\) the density of defect states. Since the two-level model only deals with cases at low injection densities, where the carrier concentration \((n)\) is proportional to \(J_f\),\(^\text{19}\) we are able to replace \(n\) with \(J_f\) directly obtained from experiments. \(\alpha\) and \(\beta\) can be obtained by curve fitting. Instead of \(\alpha\) and \(\beta\) themselves, we are more interested in their ratios: \(\gamma = \beta/\alpha = \gamma_i N_i / W_i = C_i / W_i\). Taking into account the two factors, namely, individual defect capture rate and density of defect states, the \(C_i\) in the numerator describes the capability of capturing the carriers for all PDs in the active region; thus,
it can be recognized as the defect capture coefficient—the counterpart of $A$ in the two-level model. Since the $W_r$ in the denominator remains constant with ageing, $\chi$, which can be obtained by curve fitting, is proportional to $c_t$. A fitting curve for the rising section via the two-level model is illustrated in Fig. 2(e) using a red line. We plot the curve of $\chi$ vs $J_f$ in Fig. 2(d) using a red line. The trend of $\chi$ highly resembles that of $A$, the green line in Fig. 2(d), for it also exhibits a rapid increase before $\sim 100$ h followed by a slower one. This indicates that the defect capture coefficient increases with ageing, from the point of view of the two-level model, whereas it has not been clear which factors of $c_t$ shift with ageing. From the inset of Fig. 2(d), it can be seen that there is no evident shift in $\alpha$ (the ratio between the largest and smallest $\alpha$ values is only $\sim 1.3$, while that for $\chi$ is $\sim 8.6$), the $\gamma_t$ and $W_r W_t$ of which are its denominator and numerator, respectively. The two recombination rates in the numerator should remain constant during ageing, as mentioned previously, therefore, so does the denominator $-\gamma_t$. Thus, the increase in $N_t$—the PD density—contributes markedly to the increase in defect capture coefficient. The result is a fulfillment to that obtained from the ABC model, for it has further proved that PD growth is the reason for the increase in the intensity of SRH recombination during ageing.

What could be studied from falling segments of EQE curves is the mitigation of the localization effect caused by the modification of localization centers during ageing, which was revealed previously by ELs in Fig. 1(c). The EQE curve at 0 h shows two portions at the falling segment—it decreases more rapidly at the initial part of the droop, where $I_f$ ranges from 10 to 100 mA ($1-10$ A/cm$^2$), but slows down at higher currents. Such a two-step droop has been studied both experimentally and theoretically by Wang and co-workers, who ascribe the rapid droop near the droop onset and the further slow droop to carrier delocalization and other mechanisms such as Auger or carrier leakage.$^{20,21}$ This theory has been supported by several successive works of other research groups and has proved to validate both EL and PL.$^{22,23}$ When carriers are localized, they participate in the band edge emission, increasing the EQE. As the injection density increases, another process, namely, carrier delocalization, occurs; that is, at higher $J_f$ values, localization centers are saturated and no longer capable of holding those newly injected carriers, giving them more chances to be captured by nonradiative recombination centers. Carrier localization and delocalization always come in pairs, and will in sequence cause the EQE to increase at moderate $J_f$ values, then to decrease markedly when $J_f$ is slightly higher than the droop.
The higher the intensity of carrier localization, as illustrated using the black line of 0 h in Fig. 2(b).

As a result, the initial droop rate \( \xi \), defined in Eq. (3), is a measure of the degree of carrier localization/delocalization. The higher the \( \xi \), the higher the intensity of carrier localization.

\[
\xi = \frac{\eta_{\text{max}} - \eta_{\text{i}}}{I_{\text{onset}} \eta_{\text{max}}}
\]

In Eq. (3), \( \eta_{\text{max}} \) and \( \eta_{\text{i}} \) respectively denote the EQE maximum and EQE at current \( I \), which is set at a value 10 mA higher than the onset current \( I_{\text{onset}} \). The plot of \( \xi \) vs ageing time is shown in Fig. 2(f). One could find a similar but reversed trend with \( A \) in the ABC model and \( \chi \) in the two-level model, as shown in Fig. 2(d). The fact that \( \xi \) decreases with ageing time indicates the diminishing carrier localization effect during ageing. In addition, within the first 100 h, both the formation of PDs and the reduction in the number of localization centers are faster than in successive time periods. After 100 h, the whole device becomes stable, as reflected from the obtained \( \xi, \chi, \), and \( A \). The result of the reduction in carrier localization is consistent with that obtained from the ELs. The blue shifts and linewidth expansion in the ELs indicate the removal of localization centers, while the changes in the falling sections of EQE curves confirm it and further reveal the evolution in more detail. In addition, we also compare the \( I-V \) curves tested before and during ageing, which are plotted in Fig. 3(a). During ageing, the \( I-V \) curve shifts towards high voltages, especially at lower currents.

As plotted in Fig. 3(b), forward voltages at low currents generally increase with ageing time, indicating a decrease in conductivity as well as in carrier mobility, which could be interpreted as the result of PD formation, since PDs serve as “traps” for carriers.

In summary, for InGaN green LEDs, the injection current modifies the localization centers, and at the same time reinforce the PD growth, when samples are subject to a short-term current-stress ageing, resulting in severer luminous intensity decay. The removal of localization centers by modifying the microstructure of localization centers and the proliferation of PDs should be interconnected. None of these two processes are desirable because they all give rise to efficiency decay.

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