Modelling of proton irradiated GaN-based high-power white light-emitting diodes

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We report the effects of high-energy (23 GeV) proton irradiation at large fluences on packaged high-power GaN-based white light-emitting diodes with YAG:Ce phosphors. From optical and electrical measurements, we assume that proton irradiation degrades only the GaN LED die up to fluences of at least 2 × 10^{14} p/cm², and we demonstrate that it produces nonradiative recombination centres which increase the leakage current and diminish the carrier density in the quantum wells and hence the output optical power. We also propose for the first time a model correlating optical and electrical degradation induced by radiation. © 2018 The Japan Society of Applied Physics

CERN comprises more than 45 km of particle accelerator tunnels, where lighting is today provided by fluorescent tubes with wire-wound ballasts, which are now obsolete and have to be replaced by more efficient LED-based systems. 1) Considering that thousands of luminaires (amounting to millions of LED chips) will be installed within the next years at CERN in highly radioactive areas (annual 1 MeV neutron equivalent fluence > 5 × 10^{12} n/cm² and absorbed dose > 10^3 Gy), 2,3) a thorough understanding of radiation effects on packaged high-power white GaN LEDs is therefore required. Several works in the past 20 years have investigated radiation damage on GaN LEDs; 4–21 displacement damage was shown to be the dominant degradation mechanism, decreasing light output and increasing forward leakage current due to the higher defect density. Nonetheless, only two works have so far considered phosphor-converted white LEDs: Ref. 22, in which however only low-power devices were tested and modest fluences were achieved (with no damage detected), and Ref. 23, in which γ-rays were used as radiation source; however, γ-rays are not representative of the CERN tunnel radiation environment, 3) and are not effective in inducing displacement damage (degradation in LED dies was limited even at 1 MGy). Reference 23 is also the only work testing LEDs having output powers > 1 W; to the best of our knowledge, all other works consider mid/low-power blue/green/UV devices, which were potentially manufactured using methods and technologies different from those of current high-power LEDs. Motivated by the lack of reports on packaged high-power white GaN LEDs under high-fluence particle bombardment, we have performed a dedicated irradiation campaign at CERN using 23 GeV protons on high-power white LEDs (Nichia NVSL119CT).

Nichia NVSL119CT white LEDs contain a blue emitting GaN LED with a 2 mm² die area having a flip-chip structure with patterned sapphire substrate. 24) The GaN die is assumed to be grown on the c-plane and to comprise: i) a highly doped p-GaN layer; ii) a highly doped p-AlGaN electron blocking layer; iii) a weakly n-doped GaN/InGaN multi-quantum-well (MQW) region; iv) a highly doped n-GaN layer. A reflective Ag/Ti/Au mirror layer is assumed to be deposited at the p-side contacts for improving light extraction; the GaN die is mounted on an AlN substrate via gold stud bumps. 24) We assume that white light conversion is achieved via YAG:Ce phosphors. The LED is encapsulated in a 3535 package, comprising a silicon Zener diode for ESD protection and a silicone (polysiloxane) lens for primary optics. The selected LED has a correlated colour temperature of 4000 K and a colour rendering index of 70.

In a packaged white LED, reduction of optical performance due to radiation can occur as a consequence of: i) displacement damage 7) in the blue GaN LED die; colour centre formation in ii) the sapphire substrate 25) and iii) the YAG:Ce phosphor coating layer; 26) iv) absorption band formation (due to molecular bond scissions) in the polysiloxane lens. 27) From previous results in literature and non-ionising energy loss (NIEL) calculations in GaN, 5,9) damage in GaN LEDs under 23 GeV proton irradiation is expected to start after equivalent fluences comprised between 10^{12} and 10^{13} p/cm². YAG:Ce phosphors are reported to be more resistant, with damage occurring only after 5 × 10^{14} p/cm² under 23 GeV proton bombardment. 26) Sapphire and polysiloxane degrade after even higher fluences. 25,27)

Sample irradiation was performed at IRRAD facility (CERN) using 23 GeV protons at fluences of 10^{13}, 10^{14}, and 2 × 10^{14} p/cm². Three LEDs were irradiated per each fluence. The LEDs were not powered during irradiation. Irradiation was done at room temperature (RT) and atmospheric pressure. Electrical and optical measurements were performed before and after irradiation, including light–current (L–I), electroluminescence (EL), RT and temperature-dependent current–voltage (I–V) measurements. Optical measures were taken with a FEASA LED spectrometer with an 1° mobile integrating sphere; a Keithley 2410 source-meter unit was used for current/voltage supply and measurement. A Binder MKFT climatic chamber was used for temperature dependent I–V measures. Equivalent results have been obtained for all LEDs irradiated to the same fluence; for the sake of brevity only one representative sample per fluence is presented.

The L–I curves before and after irradiation are shown in Fig. 1(a). The emitted power decreases with increasing fluence, with the highest reduction occurring at lower currents, as already noted in literature. 18–21) By interpolation of experimental data, a 50% reduction in light output at drive currents I > 0.2 A occurs at fluences corresponding to radiation exposure of more than 5 years in a typical segment of CERN tunnel complex. EL measurements are taken at the same currents of L–I measures. Figure 1(b) presents the EL...
Figure 1(c) shows the RT $I$–$V$ curves before and after irradiation. Both before and after radiation exposure, leakage current attributed to thermally assisted multi-step tunneling\cite{29} dominates in the low voltage bias region ($V \lesssim 1.5$ V), with larger currents after increasing fluences (due to increased density of defects acting as stepping-stones for tunnelling and hopping). At higher voltages ($V \gtrsim 1.5$ V), the $I$–$V$ curve before irradiation is characterized by two distinct current regimes: i) for $1.5 < V < 2.3$ V (Region A), leakage current due to nonradiative recombinations (as suggested by the invariability of the ideality factor $\eta$ in $I$–$V$ curves in the temperature range 203–303 K, see the online supplementary data at http://stacks.iop.org/JJAP/57/080304/mmedia) occurring at impurities and growth-related defects\cite{30–32} is the dominant conduction mechanism; ii) for $V > 2.35$ V (Region B), radiative recombination in quantum wells (QWs) dominates the $I$–$V$ curve. For $V \gtrsim 2.7$ V, the $I$–$V$ curve bends because of the LED series resistance. After irradiation, conduction above $\sim 1.5$ V consists of a single current regime, visible in the constant slope of the $I$–$V$ curves \cite{[32]} from $\sim 1.5$ to $\sim 2.6$ V; for $V \gtrsim 2.6$ V, the curves bend again due to the LED series resistance. The ideality factor of post-irradiation $I$–$V$ curves differs from those of the two regimes before irradiation, meaning that the conduction mechanism promoted by radiation-induced defects is different from the pre-irradiation case and suggesting that radiation-induced defects (predominantly Frenkel pairs of Ga and N atoms and complexes thereof)\cite{7} are distinct from growth defects. Since no segment with the same slope of Region B is found after irradiation, the increase in forward current must be due to radiation-induced leakage current which always dominates over the current produced by radiative recombinations. The magnitude of this leakage current increases with radiation, despite no changes in ideality factor being found: this indicates that higher radiation fluxes increase the density of defects responsible for the leakage current, but the underlying physical leakage process remains the same. The radiation-induced leakage current causes a decrease in the carrier density in QWs (hence reducing the light output). In principle, such leakage current can be due to tunnelling or Shockley–Read–Hall (SRH) recombinations at radiation-induced defects;\cite{33,34} discrimination between the two phenomena is made by studying the temperature sensitivity of the ideality factor of the $I$–$V$ curves, which is constantly close to 2 for SRH recombinations, or changes following a $1/k_BT$ law in case of tunnelling\cite{33,34} ($k_B$ is the Boltzmann constant, $T$ the absolute temperature). Temperature dependent $I$–$V$ measurements have been done in the range 203–303 K (see the online supplementary data at http://stacks.iop.org/JJAP/57/080304/mmedia) and the ideality factor of irradiated LEDs was close to 2 in the interval 1.8–2.6 V at any fluence and temperature (average value at 303 K: 1.95; absolute max. variation in the range 203–303 K: $\lesssim 12\%$). This indicates that radiation-induced defects causing leakage current are SRH nonradiative recombination centres (NRCs). The increase in tunnelling and hopping at low forward bias suggests that radiation introduces defects throughout the entire active region and cladding layers, however recombinations at radiation-induced NRCs are assumed to occur only in QWs, where carrier density is sufficiently high to justify such large increase in leakage current at moderate-to-high biases.
Based on these results, we propose for the first time a model to quantitatively correlate the optical and electrical degradation upon irradiation of MQW GaN LEDs. As a preliminary step, we introduce the model before irradiation, shown in Fig. 2(a), which has been already proposed to account for the nonidealities of real GaN LEDs.32,35,36) Diode $D_p$ represents the leakage current due to nonradiative recombinations occurring at sub-turn on voltages (i.e., Region A), diode $D_M$ represents radiative and nonradiative recombinations occurring in QWs (i.e., Region B), the shunt resistance $R_{sh}$ accounts for thermally assisted multi-step tunnelling leakage current at low voltage forward bias, and $R_Q$ accounts for the LED series resistance. The $I$–$V$ relation is

$$I = I_{M} + I_{p} + \frac{V'}{R_{sh}},$$

where $q$ is the absolute electron charge and $V' = V - R_{Q}$. Each $I$–$V$ curve before irradiation was fitted to model (1) considering $I_{M}$, $I_{p}$, $\eta_{M}$, $\eta_{p}$, $R_{sh}$, and $R_{Q}$ as fit variables; the fitting is done by solving an optimization problem via Nelder–Mead simplex algorithm37) for $V > 2.3$ V (to provide better accuracy in the range where light emission occurs). Before irradiation, since light emission occurs only in Region B, and since current therein is dominated by $I_{M}$, $R_{M}$, and $R_{sh}$ as obtained curves are in good agreement with their components: the ABC model35) relating the carrier density $n$ in QWs and the current $I_{QW}$ entering into them:

$$\frac{I_{QW}}{qV_{act}} = An + Bn^2,$$

where $V_{act}$ is the volume of the QWs, $A$ and $B$ are respectively the SRH and radiative recombination rates. At lower values of $n$, the nonradiative term prevails: $An \gg Bn^2$, so that $I_{QW} \propto An$ and $L \propto I_{QW}^2$. At higher values of $I_{QW}$, $Bn^2 \gg An$ so that $I_{QW} \propto Bn^2$ and $L \propto I_{QW}$. According to model (2), we deduce that $I_{QW} > 1$ mA is the condition to shift from predominance of nonradiative to radiative recombinations in QWs. Based on these facts, we model the relation $L = f(I_{M})$ as

$$L = \begin{cases} C_1 \cdot I_{M}^2 + C_2 \cdot I_{M} & I_{M} < 1 \text{ mA} \\ D_1 \cdot I_{M} + D_2 & I_{M} > 1 \text{ mA} \end{cases},$$

Parameters $C_1$ and $D_1$ are obtained by fitting experimental pre-irradiation $L$–$I$ curves to model (3). An example of fitted $L$–$I$ curve before irradiation is shown in Fig. 1(a); a good overall agreement with experimental data is achieved at any current.

After irradiation, we model the effect of radiation-induced NRCs in QWs through an additional parasitic diode $D_{rad}$ [see Fig. 2(b)]. In this model, the physical interpretation of $R_{S}$, $R_{sh}$, and $D_{p}$ remains the same as before irradiation. $D_{M}$ accounts for radiative transitions and nonradiative recombinations occurring in the QWs at defects existing prior to irradiation, but it does not represent any longer the overall current entering into QWs; in fact, $I_{QW} = I_{M} + I_{p}$ after irradiation. The $I$–$V$ characteristic is therefore:

$$I = I_{M} + I_{p} + \frac{V'}{R_{sh}} + I_{rad} \left( \exp \left( \frac{qV'}{\eta_{rad}kT} \right) - 1 \right),$$

where $I_{M}$ and $I_{p}$ have the same expression as before irradiation. The effect of radiation-induced NRCs in QWs are included in model (2) as follows:

$$\frac{I_{QW}}{qV_{act}} = \frac{An + Bn^2 + F(n)}{\alpha_{M} + \alpha_{act}},$$

where the density-dependent term $F(n)$ accounts for recombinations at these NRCs; $F(n)$ increases with increasing fluence. From (5), we see that the relation between $n$ and $I_{M}$ remains the same as before irradiation [i.e., $I_{M} = qV_{act}(An + Bn^2)$], and since $A$ and $B$ should not be affected by irradiation, the relation between $L$ and $I_{M}$ (and between $L$ and $n$) does not vary after irradiation, and the same relation (3) is expected to hold with the same pre-irradiation parameters.

The $I$–$V$ curve of each irradiated LED was fitted to model (4) using Nelder–Mead simplex algorithm; the fit was again performed for $V > 2.3$ V. $R_{S}$, $I_{act}$, $\eta_{rad}$, $I_{p}$, and $R_{sh}$ are considered as fit variables; conversely $\eta_{M}$, $I_{M}$, and $\eta_{p}$ are regarded as constant parameters and set equal to their corresponding pre-irradiation values. $\eta_{M}$ is assumed not to change because the conduction mechanism associated to radiative and nonradiative transitions at pre-existing defects in QWs should not be affected by radiation. Similarly, recombination at growth-related defects at sub-turn on voltages should not vary upon irradiation, because the density of these defects is not increased by irradiation, so that the parameters of $D_{p}$ are kept constant. After performing the fit, the obtained values of $I_{M}$ are used to compute the light output of irradiated LEDs via Eq. (3). The $I$–$V$ curves thus computed are shown in Fig. 1(a); a very good agreement between experimental and computed data is obtained at any current and fluence. The fitted $I$–$V$ curves after irradiation are shown in Figs. 3(a)–3(c), together with their components: the fitted curves are in good agreement with the experimental ones in the entire voltage range despite the fit was done in a limited interval. The ratio $I_{M}/I_{rad}$ [see Figs. 3(a)–3(c)] reflects the ratio of radiative to nonradiative
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The reduction of carrier density in QWs upon irradiation predicted by (5) is also confirmed by the lack of peak wavelength blueshift which normally occurs in GaN LEDs grown on a polar plane at high currents36 [see Figs. 3(d)–3(f), showing the peak wavelength \( \lambda_{\text{peak}} \) before and after each fluence]; indeed, the magnitude of blueshift monotonically decreases with increasing fluence. Figures 3(d)–3(f) also present the peak emission wavelengths computed via model (4); these values are obtained as follows: first, the relation \( \lambda_{\text{peak}} = f(I_M) \) is obtained for pre-irradiated samples by interpolation of experimental values assuming \( I \approx I_M \); then, the values after irradiation of \( \lambda_{\text{peak}} \) at different currents \( I \) are computed through the same relation \( \lambda_{\text{peak}} = f(I_M) \), using the values of \( I_M \) computed through (4) corresponding to the chosen values of \( I \). A good agreement between experimental and computed data is achieved at any current and fluence, except for currents \( \geq 200 \text{ mA} \) at \( 10^{14} \text{ p/cm}^2 \) and \( 2 \times 10^{14} \text{ p/cm}^2 \). The reason of this discrepancy is that relation \( \lambda_{\text{peak}} = f(I_M) \), obtained from pre-irradiated samples, does not account for the increased junction temperature after fluences \( \geq 10^{14} \text{ p/cm}^2 \) at high currents as after irradiation the same values of \( I_M \) are obtained at higher injected currents \( I \) (i.e., at higher junction temperatures, so that \( \lambda_{\text{peak}} \) redshifts due to band-gap shrinkage).

In summary, packaged white high-power LEDs were tested with 23 GeV proton irradiation at fluences of \( 10^{13} \), \( 10^{14} \), and \( 2 \times 10^{14} \text{ p/cm}^2 \). Measurements show no changes in the shape of EL spectra in the range 380–780 nm at any considered fluence, thus leading us to believe that radiation degrades only the GaN LED die. By performing \( L-I \) and \( I-V-T \) measurements, it is shown that proton irradiation introduces NRCs in the QWs decreasing the carrier density therein to the detriment of radiative transitions, and increasing the leakage current. We also propose for the first time a model capable of effectively correlating degradation of optical and electrical quantities and of predicting the emitted light (including its spectral features) of irradiated MQW GaN LEDs.
