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Generated carrier dynamics in V-pit enhanced InGaN/GaN light emitting diode

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Abstract

We investigate the effects of V-pits on the optical properties of a state-of-the art highly efficient, blue InGaN/GaN multi-quantum-well (MQW) light emitting diode (LED) with high internal quantum efficiency (IQE) of > 80%. The LED is structurally enhanced by incorporating pre-MQW InGaN strain-relief layer with low InN content and patterned sapphire substrate. For comparison, a conventional (unenhanced) InGaN/GaN MQW LED (with IQE of 46%) grown under similar conditions was subjected to the same measurements. Scanning transmission electron microscopy (STEM) reveals the absence of V-pits in the unenhanced LED, whereas in the enhanced LED, V-pits with $\{10\bar{1}1\}$ facets, emerging from threading dislocations (TDs) were prominent. Cathodoluminescence mapping reveals the luminescence properties near the V-pits, showing that the formation of V-pit defects can

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3 encourage the growth of defect-neutralizing barriers around TD defect states. The diminished
4 contribution of TDs in the MQWs allows indium-rich localization sites to act as efficient
5 recombination centers. Photoluminescence and time-resolved spectroscopy measurements suggest
6 that the V-pits play a significant role in the generated carrier rate and droop mechanism, showing
7 that the quantum confined Stark effect is suppressed at low generated carrier density, after which
8 the carrier dynamics and droop are governed by the carrier overflow effect.
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17 **Keywords** InGaN, efficiency droop, light emitting diode, carrier dynamics, time-resolved
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3 Blue light emitting diodes (LEDs) based on III-nitrides materials are distinguished by their structural
4 and mechanical robustness and their inherently efficient radiative recombination rates.¹⁻² At high
5 carrier injection rates, however, InGaN LEDs suffer from an efficiency droop,²⁻¹⁰ which limits their
6 performance. According to the prevalent consensus, Auger recombination is the cause of the
7 droop.⁶⁻⁸ However, some researchers have also attributed this droop to the presence of polarization
8 fields in the active layers, which facilitate electron leakage into the p-GaN layer.^{9, 11-12} In fact, it has
9 been suggested that these effects might not be mutually exclusive.¹⁰ To mitigate the deleterious
10 effects of the droop, researchers have experimented on several structural improvements. One of the
11 most prominent efforts focused on a patterned sapphire substrate (PSS) that results in stress
12 relaxation of the GaN epilayers and the reduction of TD density, leading to efficiency
13 improvement.¹³⁻¹⁶ Other approaches, based on inclusion of p-AlGaN¹⁷⁻¹⁸ or p-InGaN/AlGaN¹⁹
14 electron blocking layers (EBL) above the multi-quantum-well (MQW) LED structure, were found to
15 enhance efficiency. Additionally, incorporation of InGaN/GaN strain-relief layers, such as strained-
16 layer superlattices (SLSs) or low InN content layers, have been explored as a means to increase
17 InGaN LED efficiency by suppressing built-in polarization fields in the MQW region.²⁰⁻²² SLS layers
18 have previously been used to regulate the growth of thin quantum wells in V-pits with characteristic
19 $\{10\bar{1}1\}$ facets.²³⁻²⁵ However, the effects of V-pits on the carrier dynamics and droop mechanism in
20 III-nitride LEDs are presently not fully understood.

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41 In this work, we show the optical properties of the LED structure near such V-pits and the
42 effect of generated carriers on the optical efficiency and droop phenomenon. For comparison, we
43 also examine a conventional MQW LED grown on a flat substrate without the strain relief layer.
44 Thus, the present study advances the current understanding of the carrier dynamics and droop
45 effects in LEDs.

46 47 48 49 50 51 52 **EXPERIMENTAL DETAILS** 53 54 55 56 57 58 59 60

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3 Two blue-emitting $\text{In}_x\text{Ga}_{1-x}\text{N}/\text{GaN}$ LED structures (nominal $x \approx 0.15$) were prepared by
4 metalorganic chemical vapor deposition (MOCVD). The structurally enhanced LED sample (denoted
5 as LED1) was grown on PSS (lens-shaped patterns of $\sim 2 \mu\text{m}$ diameter) with a low InN content strain-
6 relief layer and an EBL, whereas LED2 sample was grown as a conventional LED structure on a planar
7 sapphire substrate without a strain-relief layer or EBL. We used trimethyl-indium (TMIn), trimethyl-
8 gallium (TMGa), trimethyl-aluminum (TMAI) and NH_3 . Both LED structures consisted of a low-
9 temperature GaN buffer layer overgrown on the substrates, followed by an undoped GaN layer of 3
10 μm thickness. In the next step, a 3- μm thick n-GaN layer was grown, followed by an 8-period
11 InGaN/GaN (3 nm/8 nm) MQW active layer capped by a p-GaN layer. In LED1, a strain-relief layer
12 was inserted between the n-GaN layer and the InGaN/GaN MQW active region. A p-AlGaIn EBL was
13 sandwiched between the p-GaN layer and the MQWs of LED1. For I-V and electroluminescence (EL)
14 characterization, the LEDs were fabricated by inductively coupled plasma (ICP) etching to expose the
15 n-GaN layer. Prior to ICP etching, a 500 nm SiO_2 protective layer was grown on part of the p-GaN
16 layer. This SiO_2 layer was then after, etched away using buffered oxide etchant (BOE) to expose the
17 p-GaN layer. Ni/Au (5/5 nm) current spreading layer was deposited on the p-GaN layers, following
18 which Au (150 nm) and Ti/Al/Ni/Au (10/100/30/100 nm) electrodes were subsequently deposited on
19 the exposed p-GaN and n-GaN layers, respectively. A Keithley DC power supply was used as the
20 voltage source for IV measurements, and ReRa solutions Tracer IV-curve software was used for data
21 acquisition (Figure S5, supporting information).
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43 The LED samples were prepared for scanning transmission electron microscopy (STEM) and
44 high angle annular dark field-scanning TEM images (HAADF-STEM) using an FEI Quanta 3D focused
45 Ion Beam (FIB)-Scanning Electron Microscope (SEM). The HAADF-STEM images were acquired using a
46 Cs-Probe Corrected FEI Titan, operated at an acceleration voltage of 300 kV. We estimated the V-pit
47 density after etching the p-layer and EBL by FIB-SEM. Cathodoluminescence (CL) mapping was
48 acquired at room temperature (RT) using an FEI Sirion 200 FEGSEM attached to monochromator
49 with 400 l/mm grating.²⁶ The electron beam energy was fixed at 10 keV for CL mapping. For power-
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3 dependent RT-photoluminescence (PL) measurements, the second harmonic line (400 nm) of an
4 ultrafast (150 fs) Ti:Sapphire laser (76 MHz) was used. For temporally resolved RT-PL (TRPL)
5 measurements, an APE GmbH pulse picker was used to reduce the pulse frequency to 1 MHz. The
6 diameter of the incident beam was $\sim 60 \mu\text{m}$. A charge-coupled device camera attached to a
7 Hamamatsu single-sweep streak camera was used to acquire both the temporal and time integrated
8 responses. The samples were mounted in a closed-cycle helium cryostat for low temperature (5 K)
9 measurements.
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18 RESULTS AND DISCUSSION

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21 Figure 1(a) shows the cross-sectional STEM images of the LED1. We observe a TD defect
22 (circled area) emerge from the center of the V-pit defect into the p-GaN layer of the sample. SEM
23 images reveal that the average V-pit density is $\approx 1.5 \times 10^8 \text{ cm}^{-2}$ as shown in Figure S2 of the
24 Supporting Information.²⁷ The facets of the V-pit walls are separated by a $\approx 63^\circ$ angle, which
25 coincides with the angle separating the $\{10\bar{1}1\}$ group of planes of hexagonal InGaN structures.²⁸⁻²⁹
26 The STEM image shows that the V-pit walls are characterized by quantum well and barrier thinning,
27 in line with previous observations.²³⁻²⁴ It is well known that different planer facets of III-nitride based
28 crystal lattices have different surface energies,³⁰⁻³¹ which can lead to strong dependence of adatom
29 kinetics on the crystal plane orientation.³² Indeed, it was shown by Hangleiter, *et al.*²⁵ demonstrated
30 that the In growth rate along the semi-polar plane is slow, which would explain the MQW thinning.
31 V-pits are not observed in LED2, as shown in Figure 1(b), where TDs can be seen cleaving through its
32 MQWs.
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47 We investigate the detailed emission spectrum of LED1 to study the effect of the V-pits. We
48 plot the mean CL energy spectrum of LED1 emission (Figure 2(a)), showing that the main energy
49 peak is centered at $\sim 2.71 \text{ eV}$. The CL intensity maps corresponding to the color shaded energy
50 regions are shown alongside the spectrum. The low energy InGaN shoulder, located in the 2.35–2.70
51 eV range, corresponds to the brighter areas on the CL map in Figure 2(b), while the dark spots are
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3 signatures of the V-pits. The shoulder can be due to InN-rich fluctuations inside the MQWs. The
4 bright regions shown in Figure 2(c) correspond to the MQW emission in the 2.75–2.90 eV range. In
5 the same figure, the V-pit spots become smaller due to reduced InN content in the areas in the
6 immediate vicinity of V-pits. The high energy shoulder located between 3.05 and 3.25 eV represents
7 the emission from the V-pits (bright spots in the CL map of Figure 2(d)). It should be noted that the
8 low InN layer is also located within this range, however its intensity is low enough to provide
9 contrast for the inner walls of the V-pits due to the GaN quantum barrier above this layer (Figure S3,
10 supporting material).²⁷ Figure 2(e) shows the CL map related to emission near the GaN bandedge
11 (located at 3.35–3.50 eV), indicating that the V-pits become sparse, suggesting that p-GaN, the V-pit
12 walls and the preceding GaN layers may have contributed to this peak.
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24 In Figure 3(a) and (b), we compare the hyperspectral CL intensity maps of LED1 and LED2,
25 respectively, integrated over the 2.4–2.9 eV energy range. Unlike in LED1, we find no evidence of V-
26 pits in LED2 and large sections of low CL intensity are shown (Figure 3(b)) overshadowing patches
27 of high intensity regions. Figure 3(c) and (d) show the corresponding CL spectra of the regions
28 annotated as A and B, in the CL maps (Figure 3(a) and 3(b), respectively). A high energy shoulder
29 peak at ≈ 3.03 eV that is related to well-barrier intermixing (Figure 2(d)) in LED1 is absent from the
30 LED2 emission spectrum and no significant spectral distinctions exist between the dark and bright
31 regions due to the absence of such V-pits. Figure 3(e) and (f) show the correlations between the
32 LED centroid energy and the CL intensity (integrated over the 2.4–3.2 eV range). Figure 3(e)
33 indicates a clear inverse correlation between the peak energy and CL intensity in LED1. This
34 correlation leads us to deduce that the V-pits, corresponding to the high energy shoulder (Figure
35 2(d) and Figure 3(c)), act as dislocation terminals. The walls of V-pits are known to act as TD
36 passivating barriers, due to thinner MQW walls on the facets of the pits.^{21, 25, 33} Well-barrier
37 intermixing inside of the v-pits may also play a role in the TD passivation according to Pereira, *et al.*
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3 higher intensity (represented in Figure 2(c)) relative to the high energy shoulder, suggesting a
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5 higher efficiency of radiative recombination processes within InN-rich potentials in the MQWs. In
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7 contrast, Figure 3(f) shows a positive correlation between mean peak energy and CL intensity in
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9 LED2, implying that CL quenching occurs around InN-rich sites. This finding indicates that, in the
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11 absence of TD passivating V-pits, InN tends to accumulate near TD sites.³⁴⁻³⁵
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14 We investigate the radiative recombination efficiency of the carriers and the nature of the
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16 droop in the enhanced LED (LED1) compared to the conventional one (LED2), by conducting power-
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18 dependent PL measurements at RT and interpreting the results using the Shockley-Read-Hall (SRH)
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20 model.³⁶⁻³⁷ The rate equation of generated carriers, G ($cm^{-3}s^{-1}$), in steady state is given by:
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$$22 \quad G = An + Bn^2 + Cn^3 \quad (1)$$

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24 where A, B and C are the coefficients of (non-radiative) Shockley-Read-Hall (SRH), radiative and
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26 Auger recombination, respectively. $G(cm^{-3}s^{-1})$ can then be estimated from the average excitation
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28 power value (P_{av}) as follows:
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$$31 \quad G(P) = \frac{P_{av} \times \tau_D \times \alpha \times (1-R)}{\tau_w \times A \times h\nu \times q} \quad (2)$$

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33 where α is the absorption coefficient of InGaN, linearly extrapolated from the values for InN and
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35 GaN (SRH Method, supporting information),²⁷ R is the reflectivity of the GaN surface at 3.1 eV
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37 (10%),³⁸ τ_D is the pulse duration, τ_w is the pulse width, A denotes the area of the incident excitation
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39 beam, $h\nu$ is the laser photon energy, and q represents the elementary charge. It follows from Eq. (1)
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41 that the observed integrated luminescence intensity at RT, $I(P)$, of the samples is represented by the
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43 following equation:
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$$48 \quad I(P) = kBn^2 \quad (3)$$

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50 where k is a constant related to the product of the spectrograph's collection efficiency and light
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52 extraction efficiency of the LEDs. Combining Eq. (1), (2) and (3), the LED internal quantum
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54 efficiencies (IQE), $\eta_{IQE}(P)$, can be determined by the ratio:
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$$\eta_{IQE}(P) = \frac{Bn^2}{G(P)} = \frac{I(P)}{k G(P)} \quad (4)$$

The value of k is estimated using the following steps. First, the number of parameters is reduced by restricting the analysis to low carrier generation rates ($G < 10^{31} \text{ cm}^{-3} \text{ s}^{-1}$) where Auger recombination is negligible and thus allowing the third term in Eq. (1) to be eliminated.³⁹ Substituting for n from Eq. (3) gives the new equation, $G = \left(A/\sqrt{kB} \right) I(P)^{\frac{1}{2}} + \left(1/k \right) I(P)$, which was then fitted into a plot of $G(P)$ versus the integrated luminescence intensity, $I(P)$, where A , B and k were treated as constant fitting parameters (Figure 3S, Supporting Information).²⁷ Finally, we estimated the value of $\eta_{IQE}(P)$ by substituting k into Eq. (4).

Figure 4(a) shows the carrier generation rate, G , dependence (in log scale) of IQE for both LEDs, excited by 400 nm (below the GaN bandgap). To interpret the IQE behavior of both LEDs, we define three prominent regions of interest (annotated as RI, RII and RIII Figure 4). Region RI follows the linear dependence of the SRH recombination (non-radiative) rate (An) with G . In the range of this measurement, the RI region is observed in LED2 for $4.5 \times 10^{27} < G < 8.6 \times 10^{28} \text{ cm}^{-3} \text{ s}^{-1}$, whereas this region is not observed in the structurally enhanced LED1. Since $\eta_{IQE}(P)$ is a function of radiative recombination (Eq. (4)), a slight increase in $\eta_{IQE}(P)$ of LED2 occurs because the main LED2 peak significantly overlaps with the defect band, which increases linearly with G , at low carrier density (Figure 4(d)). Such overlap is not observed at either low or high carrier densities in LED1 (Figure 4(c)). In the region denoted as RII ($4.5 \times 10^{27} < G < 6.7 \times 10^{29} \text{ cm}^{-3} \text{ s}^{-1}$ for LED1 and $1.0 \times 10^{29} < G < 6.1 \times 10^{30} \text{ cm}^{-3} \text{ s}^{-1}$ for LED2), $\eta_{IQE}(P)$ of both LEDs increases rapidly until it reaches a maximum value. This region seems more consistent with the radiative term (Bn^2) in Eq. (1). The maximum IQE for LED1 is > 80%, compared to 46% obtained for LED2. In the region denoted as RIII, there is a saturation in radiative efficiency and the droop effect occurs, where the IQE of both LEDs starts to decline, albeit at different G values. The droop effect commences at around $G \approx 2.3 \times 10^{30} \text{ cm}^{-3} \text{ s}^{-1}$ for LED1, and at $G \approx 9.0 \times 10^{30} \text{ cm}^{-3} \text{ s}^{-1}$ for LED2, consistent with the previously reported value of $\sim 3.7 \times 10^{31} \text{ cm}^{-3} \text{ s}^{-1}$.³⁹ The commencement of the droop effect at lower generated carrier density in LED1 is most likely due to

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3 the reduction in the effective MQW volume, as a result of high v-pit defect density. A similar
4 observation was made by estimating the IQE of both LEDs by EL (Figure S6). In this case, the
5 maximum IQE values for both LEDs were consistent with the PL results (a more detailed discussion of
6 this measurement is provided in the supporting information. The internal quantum efficiency,
7 $\eta_{IQE}(P)$, eventually declined to 48% and 30% at $G \approx 5.3 \times 10^{31} \text{ cm}^{-3}\text{s}^{-1}$ for LED1 and LED2, respectively.
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9 Furthermore, the inset of Figure 4(a) shows that the efficiency droop characteristics of the two LEDs
10 behave differently as the excitation power intensity increases. The droop regime of LED2 follows a
11 convex curve, likely attributed to the dominant effect of defect-related non-radiative recombination
12 through the SRH process,⁴⁰ whereas that of LED1 follows a concave IQE curve, indicating that the
13 droop could be due to the carrier overflow mechanism. When the effect of SRH recombination is
14 significantly diminished, carrier overflow becomes the dominant source of efficiency droop.⁴⁰

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16 To further explain the droop behavior, we investigate the dependence of the peak energy of
17 both LEDs on G (Figure 4(b)). We observe a clear blue-shift of ~ 80 meV in LED1 as the excitation
18 power increases. However, the peak position of LED2 remains initially unchanged, before slightly
19 blue-shifting by 10 meV at $G \approx 6.7 \times 10^{29} \text{ cm}^{-3}\text{s}^{-1}$. To understand these distinct behaviors, the full width
20 at half maximum (FWHM) values of the emission peaks of both LEDs are plotted as a function of G
21 (Figure 4(b), inset). For LED1, the peak FWHM decreases slightly (by 5 meV) as G increases from
22 4.5×10^{27} to $3.7 \times 10^{28} \text{ cm}^{-3}\text{s}^{-1}$, before broadening by 30 meV to 155 meV at higher G values, followed
23 by an invariant response to generated carriers, starting from $G \approx 5.2 \times 10^{30} \text{ cm}^{-3}\text{s}^{-1}$. For LED2, the
24 FWHM initially decreases significantly (by 55 meV, from 165 meV to 110 meV for $4.5 \times 10^{27} < G <$
25 $1.0 \times 10^{29} \text{ cm}^{-3}\text{s}^{-1}$), before broadening by 43 meV as G increases. The FWHM narrows as the carrier
26 density increases due to the screening of strain-induced electric field (i.e., quantum confined Stark
27 effect (QCSE)) by the increasing carrier population. However, FWHM broadening accompanied by
28 blue-shifting of the peak energy as carrier density increases is caused by carrier overflow from
29 deeply localized InN-rich states to shallower states and other higher energy states. By this point, the
30 QCSE is fully suppressed by the high carrier density.⁴¹ Thus, the QCSE effect is not significant in LED1

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3 and is overcome at low G value ($10^{28} \text{ cm}^{-3}\text{s}^{-1}$). Consequently, its Auger dynamics are governed by the
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5 overflow effect. We therefore propose that, beyond the QCSE screening limit, the observed peak
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7 blue-shift in LED1 can be attributed to the carrier saturation of strong localization centers (due to
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9 indium segregation) and subsequent occupation of weak states inside the well,⁴² followed by carrier
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11 occupation of states inside the V-pits. Given that the Auger effect of LED1 is barely affected by
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13 piezoelectric polarization, carrier overflow around the V-pits is suggested as the mechanism behind
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15 the characteristic concave droop behavior observed in LED1.^{10, 40} This mechanism is illustrated in
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17 Figure 4(e). This assertion is supported by the fact that the droop effect commences at the same G
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19 value ($\approx 10^{30} \text{ cm}^{-3}\text{s}^{-1}$) that the FWHM became constant. It is also plausible to assume that excess
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21 carriers may overflow into the p-GaN region as well.⁴⁰ However this effect should not be significant,
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23 since the excitation photon energy (3.1 eV) is markedly below the AlGaIn EBL bandgap. For LED2, the
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25 limited dependence of the peak energy on carrier generation rate and the initial narrowing of its
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27 FWHM (Figure 4(b)) indicate that the carriers are already weakly confined, leaving excess carriers to
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29 contend with a nontrivial contribution from QCSE and the defect states of LED2. The FWHM of LED2
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31 initially plateaus before starting to increase at high G ($> 10^{29} \text{ cm}^{-3}\text{s}^{-1}$) values, indicating that the full
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33 screening of the QCSE in LED2 occurs at an additional order of magnitude than LED1, causing a slight
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35 blue-shift at $G \approx 6.7 \times 10^{29} \text{ cm}^{-3}\text{s}^{-1}$. Thus, LED2's Auger effect may have been affected by the
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37 continuous evolution of the electron-hole overlap ratio due to the dependence of its polarization
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39 field on injected carrier density, which explains its convex droop characteristic.¹⁰
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44 Power-dependent TRPL was carried out to confirm the contribution of radiative and non-
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46 radiative recombination in LED1 compared to LED2. Fig. 5(a) and 5(b) respectively show the power-
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48 dependent TRPL lifetimes of LED1 and LED2 taken at 5 K. LED1 exhibits a non-exponential carrier
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50 lifetime decay (a similar behavior is observed at 290 K), suggesting presence of multi-state
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52 recombination paths, which can be approximated by the bi-exponential equation:⁴³
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$$55 \text{Int}(t) = A_f e^{-t/\tau_f} + A_s e^{-t/\tau_s}, \quad (5)$$

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3 where A_f and A_s are, respectively, the fast and slow peak intensities at time $t = 0$, while τ_f and τ_s
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5 denote the decay lifetimes of the fast and slow decay components. However, LED2 exhibits a single
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7 exponential decay, suggesting that the excess carrier recombination paths in the two LEDs are
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9 different.

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11 Figure 6 shows the PL decay lifetimes as a function of G at both 5 K and 290 K for LED1 and
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13 LED2, respectively. At 5 K, the PL lifetime of the LED1 peak declines from 96 ns to 47 ns for $6 \times 10^{27} <$
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15 $G < 2.7 \times 10^{28} \text{ cm}^{-3} \text{ s}^{-1}$, after which it remains constant (Figure 6(a)). This inverse proportionality of
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17 radiative carrier lifetime to excitation carrier density implies that defect-related non-radiative
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19 recombination plays a negligible role at 5 K^{17, 44}. However, at 290 K, the PL lifetime increases with G
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21 until $4.2 \times 10^{28} \text{ cm}^{-3} \text{ s}^{-1}$, which is due to the increase in non-radiative lifetime of LED1, as shown by the
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23 radiative and non-radiative lifetime in the inset of Figure 6(a). This behavior is followed by a
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25 subsequent reduction in the PL lifetime when radiative recombination starts to dominate the
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27 recombination process due to the saturation of non-radiative defect sites.⁴⁵ This behavior confirms
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29 that the non-radiative recombination processes become influential at high temperatures only, when
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31 thermal activation contributes to the deconfinement of previously confined carriers. Nonetheless,
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33 this effect occurs at low G values only ($G < 10^{29} \text{ cm}^{-3} \text{ s}^{-1}$, IQE \ll 50%). For LED2, Figure 6(b) shows that
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35 the PL lifetime increases initially at 5 K (from 12 ns to 14 ns in the $6.0 \times 10^{27} < G < 1.8 \times 10^{28} \text{ cm}^{-3} \text{ s}^{-1}$
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37 range) before decreasing as G increases, whereas at 290 K its PL lifetime increases monotonously
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39 with G . There is a striking similarity between LED2's behavior at 5 K and that of LED1 at 290 K.
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41 Therefore, we posit that, at low excitation intensities, defect-related non-radiative recombination
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43 plays a prominent role in the recombination processes of LED2 at 5K. At RT, non-radiative processes
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45 dominate recombination rates beyond $G = 10^{29} \text{ cm}^{-3} \text{ s}^{-1}$ (inset of Figure 6(b)). This finding is also
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47 supported by I-V plots (Figure S5, supporting information),²⁷ which show that the effect of shunt
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49 resistance was less severe in LED1 than in LED2. Shunt resistance is indicative of damaged regions or
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51 surface imperfections¹⁷ which may result from dislocation defects.⁴⁶ These results confirm that the
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53 role of defect-related recombination was far less significant in LED1 than in LED2.
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CONCLUSION

We investigated the carrier dynamics of a V-pit enhanced MQW LED. At low carrier densities, the V-pits acted as TD passivating barriers, thereby permitting efficient radiative recombination in the wells. However, as carrier density increases, the reduced effective volume of the MQWs allows for an early onset of Auger phenomenon. We further show that the Auger droop effect is mainly driven by carrier overflow, rather than piezoelectric polarization or SRH defects in the V-pit enhanced LED. Lifetime measurements show that the improved efficiency of the carrier recombination processes in the structurally enhanced LED was significantly aided by the presence of V-pits, leading to dominant radiative recombination process at RT.

ASSOCIATED CONTENT

Supporting information

Estimation of p-GaN doping concentration by SIMS; details of V-pit density estimation by SEM; G(P) vs I(P) plots and fittings; I-V characteristics and peak overlap explanation between the strain relief layer and the V-pits, electroluminescence IQE, off-center cut characteristics of V-pits, and EDX compositional mapping around the V-pits . This material is available free of charge via the Internet at <http://pubs.acs.org>.

AUTHOR INFORMATION

All authors have given approval to the final version of the manuscript.

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Notes

The authors declare no competing financial interest.

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REFERENCES

1. Lester, S. D.; Ponce, F. A.; Craford, M. G.; Steigerwald, D. A., High dislocation densities in high efficiency GaN-based light-emitting diodes. *Appl. Phys. Lett.* **1995**, *66*, 1249-1251.
2. Yang, T.-J.; Shivaraman, R.; Speck, J. S.; Wu, Y.-R., The influence of random indium alloy fluctuations in indium gallium nitride quantum wells on the device behavior. *J. Appl. Phys.* **2014**, *116*, 113104.
3. Kim, A. Y.; Götz, W.; Steigerwald, D. A.; Wierer, J. J.; Gardner, N. F.; Sun, J.; Stockman, S. A.; Martin, P. S.; Krames, M. R.; Kern, R. S.; Steranka, F. M., Performance of High-Power AlInGaN Light Emitting Diodes. *physica status solidi (a)* **2001**, *188*, 15-21.
4. Takashi, M.; Motokazu, Y.; Shuji, N., Characteristics of InGaN-Based UV/Blue/Green/Amber/Red Light-Emitting Diodes. *Jpn. J. Appl. Phys.* **1999**, *38*, 3976.
5. Dong-Pyo, H.; Jong-In, S.; Dong-Soo, S.; Kyu-Sang, K., Effects of unbalanced carrier injection on the performance characteristics of InGaN light-emitting diodes. *Applied Physics Express* **2016**, *9*, 081002.
6. Shen, Y. C.; Mueller, G. O.; Watanabe, S.; Gardner, N. F.; Munkholm, A.; Krames, M. R., Auger recombination in InGaN measured by photoluminescence. *Appl. Phys. Lett.* **2007**, *91*, 141101.
7. Delaney, K. T.; Rinke, P.; Van de Walle, C. G., Auger recombination rates in nitrides from first principles. *Appl. Phys. Lett.* **2009**, *94*, 191109.
8. Iveland, J.; Martinelli, L.; Peretti, J.; Speck, J. S.; Weisbuch, C., Direct Measurement of Auger Electrons Emitted from a Semiconductor Light-Emitting Diode under Electrical Injection: Identification of the Dominant Mechanism for Efficiency Droop. *Phys. Rev. Lett.* **2013**, *110*, 177406.
9. Kim, M.-H.; Schubert, M. F.; Dai, Q.; Kim, J. K.; Schubert, E. F.; Piprek, J.; Park, Y., Origin of efficiency droop in GaN-based light-emitting diodes. *Appl. Phys. Lett.* **2007**, *91*, 183507.
10. Piprek, J.; Römer, F.; Witzigmann, B., On the uncertainty of the Auger recombination coefficient extracted from InGaN/GaN light-emitting diode efficiency droop measurements. *Appl. Phys. Lett.* **2015**, *106*, 101101.

- 1
2
3 11. Fu, H.; Lu, Z.; Zhao, Y., Analysis of low efficiency droop of semipolar InGaN quantum well
4 light-emitting diodes by modified rate equation with weak phase-space filling effect. *AIP*
5 *Advances* **2016**, *6*, 065013.
6
- 7
8 12. Ryu, H.-Y.; Shin, D.-S.; Shim, J.-I., Analysis of efficiency droop in nitride light-emitting diodes
9 by the reduced effective volume of InGaN active material. *Appl. Phys. Lett.* **2012**, *100*,
10 131109.
11
- 12
13 13. Gao, H.; Yan, F.; Zhang, Y.; Li, J.; Zeng, Y.; Wang, G., Improvement of the performance of
14 GaN-based LEDs grown on sapphire substrates patterned by wet and ICP etching. *Solid-State*
15 *Electron.* **2008**, *52*, 962-967.
16
- 17
18 14. Tadatomo, K.; Okagawa, H.; Ohuchi, Y.; Tsunekawa, T.; Jyouichi, T.; Imada, Y.; Kato, M.;
19 Kudo, H.; Taguchi, T., High Output Power InGaN Ultraviolet Light-Emitting Diodes Fabricated
20 on Patterned Substrates Using Metalorganic Vapor Phase Epitaxy. *physica status solidi (a)*
21 **2001**, *188*, 121-125.
22
- 23
24 15. Lee, K.; Lee, H.; Lee, C.-R.; Kim, J. S.; Lee, J. H.; Ryu, M.-Y.; Leem, J.-Y., Spatial emission
25 distribution of InGaN/GaN light-emitting diodes depending on the pattern structures. *Mater.*
26 *Res. Bull.* **2014**, *58*, 121-125.
27
- 28
29 16. Lee, Y.-C.; Yeh, S.-C.; Chou, Y.-Y.; Tsai, P.-J.; Pan, J.-W.; Chou, H.-M.; Hou, C.-H.; Chang, Y.-Y.;
30 Chu, M.-S.; Wu, C.-H.; Ho, C.-H., High-efficiency InGaN-based LEDs grown on patterned
31 sapphire substrates using nanoimprinting technology. *Microelectronic Engineering* **2013**,
32 *105*, 86-90.
33
- 34
35 17. Schubert, E. F., *Light-emitting diodes*. 2nd ed.; Cambridge University Press: Cambridge ; New
36 York, 2006; p x, 422 p.
37
- 38
39 18. Yu, C.-T.; Lai, W.-C.; Yen, C.-H.; Chang, S.-J., Effects of InGaN layer thickness of AlGaIn/InGaIn
40 superlattice electron blocking layer on the overall efficiency and efficiency droops of GaN-
41 based light emitting diodes. *Optics Express* **2014**, *22*, A663-A670.
42
- 43
44 19. Liu, Z.; Ma, J.; Yi, X.; Guo, E.; Wang, L.; Wang, J.; Lu, N.; Li, J.; Ferguson, I.; Melton, A., p-
45 InGaN/AlGaIn electron blocking layer for InGaN/GaN blue light-emitting diodes. *Appl. Phys.*
46 *Lett.* **2012**, *101*, 261106.
47
- 48
49 20. Ju, J.-W.; Kang, E.-S.; Kim, H.-S.; Jang, L.-W.; Ahn, H.-K.; Jeon, J.-W.; Leea, I.-H.; Baek, J. H.,
50 Metal-organic chemical vapor deposition growth of InGaN/GaN high power green light
51 emitting diode: Effects of InGaN well protection and electron reservoir layer. *J. Appl. Phys.*
52 **2007**, *102*, 053519.
53
- 54
55 21. Leem, S. J.; Shin, Y. C.; Kim, K. C.; Kim, E. H.; Sung, Y. M.; Moon, Y.; Hwang, S. M.; Kim, T. G.,
56 The effect of the low-mole InGaIn structure and InGaIn/GaN strained layer superlattices on
57 optical performance of multiple quantum well active layers. *J. Cryst. Growth* **2008**, *311*, 103-
58 106.
59
60

- 1
2
3
4 22. Davies, M. J.; Dawson, P.; Massabuau, F. C.-P.; Oliver, R. A.; Kappers, M. J.; Humphreys, C. J.,
5 The effects of Si-doped prelayers on the optical properties of InGaN/GaN single quantum
6 well structures. *Appl. Phys. Lett.* **2014**, *105*, 092106.
7
8
9 23. Chang, C.-Y.; Li, H.; Shih, Y.-T.; Lu, T.-C., Manipulation of nanoscale V-pits to optimize internal
10 quantum efficiency of InGaN multiple quantum wells. *Appl. Phys. Lett.* **2015**, *106*, 091104.
11
12 24. Tomiya, S.; Kanitani, Y.; Tanaka, S.; Ohkubo, T.; Hono, K., Atomic scale characterization of
13 GaInN/GaN multiple quantum wells in V-shaped pits. *Appl. Phys. Lett.* **2011**, *98*, 181904.
14
15
16 25. Hangleiter, A.; Hitzel, F.; Netzel, C.; Fuhrmann, D.; Rossow, U.; Ade, G.; Hinze, P.,
17 Suppression of Nonradiative Recombination by V-Shaped Pits in GaInN/GaN Quantum Wells
18 Produces a Large Increase in the Light Emission Efficiency. *Phys. Rev. Lett.* **2005**, *95*, 127402.
19
20
21 26. Paul, R. E.; Robert, W. M., Cathodoluminescence nano-characterization of semiconductors.
22 *Semicond. Sci. Tech.* **2011**, *26*, 064005.
23
24
25 27. Supporting information: V-pit density estimation by SEM, G(P) vs I(P) plots and fittings, and
26 estimation of doping p-GaN doping concentration by SIMS, I-V Characteristics.
27
28
29 28. Bessolov, V. N.; Konenkova, E. V.; Kukushkin, S. A.; Osipov, A. V.; Rodin, S. N., Semipolar
30 Gallium Nitride on Silicon: Technology and Properties. *Reviews on Advanced Materials
31 Science* **2014**, *38*, 75-93.
32
33
34 29. Vladislav, V.; Natalia, B.; Ruslan, G.; Philipp, L.; Yuri, L.; Yury, R.; Alexander, T.; Andrey, Z.;
35 Yuri, S., Nature of V-Shaped Defects in GaN. *Jpn. J. Appl. Phys.* **2013**, *52*, 08JE14.
36
37
38 30. Northrup, J. E.; Neugebauer, J., Theory of GaN(10⁻¹⁰) and (11⁻²⁰) surfaces. *Physical Review
39 B* **1996**, *53*, R10477-R10480.
40
41
42 31. Neugebauer, J., Ab initio Analysis of Surface Structure and Adatom Kinetics of Group-III
43 Nitrides. *physica status solidi (b)* **2001**, *227*, 93-114.
44
45
46 32. Girgel, I.; Edwards, P. R.; Le Boulbar, E.; Allsopp, D. W.; Martin, R. W.; Shields, P. A. In
47 *Investigation of facet-dependent InGaN growth for core-shell LEDs*, Proc. SPIE 9363, 2015; pp
48 93631V-93631V-8.
49
50
51 33. Kim, J.; Cho, Y.-H.; Ko, D.-S.; Li, X.-S.; Won, J.-Y.; Lee, E.; Park, S.-H.; Kim, J.-Y.; Kim, S.,
52 Influence of V-pits on the efficiency droop in InGaN/GaN quantum wells. *Optics Express*
53 **2014**, *22*, A857-A866.
54
55
56
57
58
59
60

- 1
2
3 34. Horton, M. K.; Rhode, S.; Sahonta, S.-L.; Kappers, M. J.; Haigh, S. J.; Pennycook, T. J.;
4 Humphreys, C. J.; Dusane, R. O.; Moram, M. A., Segregation of In to Dislocations in InGaN.
5 *Nano Letters* **2015**, *15*, 923-930.
6
7
8 35. Duxbury, N.; Bangert, U.; Dawson, P.; Thrush, E. J.; Stricht, W. V. d.; Jacobs, K.; Moerman, I.,
9 Indium segregation in InGaN quantum-well structures. *Appl. Phys. Lett.* **2000**, *76*, 1600-1602.
10
11 36. Yoshida, H.; Kuwabara, M.; Yamashita, Y.; Uchiyama, K.; Kan, H., Radiative and nonradiative
12 recombination in an ultraviolet GaN/AlGaIn multiple-quantum-well laser diode. *Appl. Phys.*
13 *Lett.* **2010**, *96*, 211122.
14
15
16 37. Dai, Q.; Schubert, M. F.; Kim, M. H.; Kim, J. K.; Schubert, E. F.; Koleske, D. D.; Crawford, M.
17 H.; Lee, S. R.; Fischer, A. J.; Thaler, G.; Banas, M. A., Internal quantum efficiency and
18 nonradiative recombination coefficient of GaInN/GaN multiple quantum wells with different
19 dislocation densities. *Appl. Phys. Lett.* **2009**, *94*, 111109.
20
21
22 38. Bloom, S.; Harbeke, G.; Meier, E.; Ortenburger, I. B., Band Structure and Reflectivity of GaN.
23 *physica status solidi (b)* **1974**, *66*, 161-168.
24
25
26 39. Xie, J.; Ni, X.; Fan, Q.; Shimada, R.; Özgür, Ü.; Morkoç, H., On the efficiency droop in InGaIn
27 multiple quantum well blue light emitting diodes and its reduction with p-doped quantum
28 well barriers. *Appl. Phys. Lett.* **2008**, *93*, 121107.
29
30
31 40. Seong, T. Y.; Han, J.; Amano, H.; Morkoc, H., III-Nitride Based Light Emitting Diodes and
32 Applications. *III-Nitride Based Light Emitting Diodes and Applications* **2013**, *126*, 171.
33
34
35 41. Feng, Z. C., *III-nitride devices and nanoengineering*. Imperial College Press: London, 2008; p
36 xiv, 462 p.
37
38
39 42. Sang-Youp, Y.; Joon Heon, K.; Mun Seok, J.; Seung-Han, P.; Jongmin, L., Power Dependent
40 Micro-Photoluminescence of Green-InGaIn/GaN Multiple Quantum Wells. *Jpn. J. Appl. Phys.*
41 **2011**, *50*, 050204.
42
43
44 43. Özgür, Ü.; Fu, Y.; Moon, Y. T.; Yun, F.; Morkoç, H.; Everitt, H. O.; Park, S. S.; Lee, K. Y., Long
45 carrier lifetimes in GaN epitaxial layers grown using TiN porous network templates. *Appl.*
46 *Phys. Lett.* **2005**, *86*, 232106.
47
48
49 44. Fukuda, M., *Optical semiconductor devices*. Wiley: New York, 1999; p xvi, 422 p.
50
51
52 45. Murotani, H.; Yamada, Y.; Honda, Y.; Amano, H., Excitation density dependence of radiative
53 and nonradiative recombination lifetimes in InGaIn/GaN multiple quantum wells. *physica*
54 *status solidi (b)* **2015**, *252*, 940-945.
55
56
57
58
59
60

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2
3 46. Zhang, Y.; Kappers, M. J.; Zhu, D.; Oehler, F.; Gao, F.; Humphreys, C. J., The effect of
4 dislocations on the efficiency of InGaN/GaN solar cells. *Solar Energy Materials and Solar Cells*
5 **2013**, *117*, 279-284.
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8 Captions

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10 Figure 1 (a) Cross-sectional STEM image of LED1's V-pits with MQW thinning on the $\{10\bar{1}1\}$ facets.
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13 (b) Cross-sectional STEM image showing LED2 MQWs with TD defects.
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16 Figure 2 (a) Mean CL emission spectrum of LED1 generated from a $10\ \mu\text{m} \times 10\ \mu\text{m}$ area
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18 hyperspectral map with (b), (c), (d) and (e) CL intensity maps corresponding to the color shaded
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20 regions marked on the spectrum.
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22 Figure 3 (a) and (b) RT-CL intensity maps of the two LEDs, (c) and (d) CL Spectra of the annotated
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24 regions on the micrographs, (e) and (f) Correlation of centroid energy (between 2.4 – 3.2 eV) vs
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26 intensity (for LED1 and LED2, respectively).
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29 Figure 4 Carrier generation rate dependence of (a) IQE (inset: IQE vs excitation power intensity
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31 (linear-scale)) and (b) peak energies (inset: FWHM vs G). PL spectra of (c) LED1 and (d) LED2 at high
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33 and low G. (e) the carrier dynamic and droop mechanism of LED1 as G increases.
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36 Figure 5 TRPL temporal response at different excitation powers (~ 0.044 – $0.76\ \text{MW}/\text{cm}^2$) of (a) LED1
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38 and (b) LED2.
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41 Figure 6 PL lifetimes of (a) LED1, and (b) LED2 as a function of G at 5K (black squares) and 290K (red
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43 circles). Insets are the radiative (blue squares) and non-radiative (magenta circles) lifetimes with
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45 respect to G at 290K.
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Generated carrier dynamics in V-pit enhanced InGaN/GaN light emitting diode

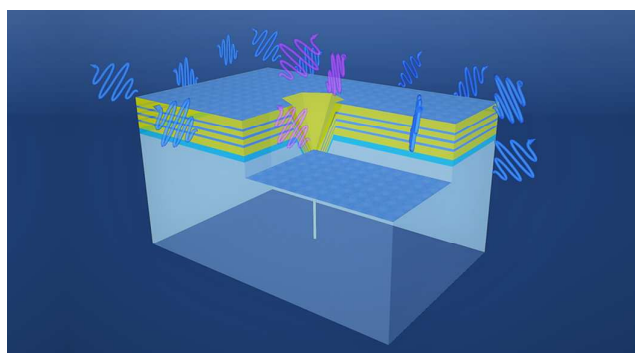
Idris A. Ajja[†]; Paul R. Edwards^{||}, Yusin Pak[†], Ermek Belekov[†]; Manuel A. Roldan[‡]; Nini Wei[‡]; Zhiqiang Liu[§], Robert W. Martin^{||} and Iman S. Roqan^{†,*}

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[‡]King Abdullah University of Science and Technology (KAUST), Imaging and Characterization Core Laboratory, Thuwal, Saudi Arabia.

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Synopsis: This graphic shows a V-pit with the different energy of photons emerging from the MQWs (low energy blue light) and the V-pits (high energy violet light)

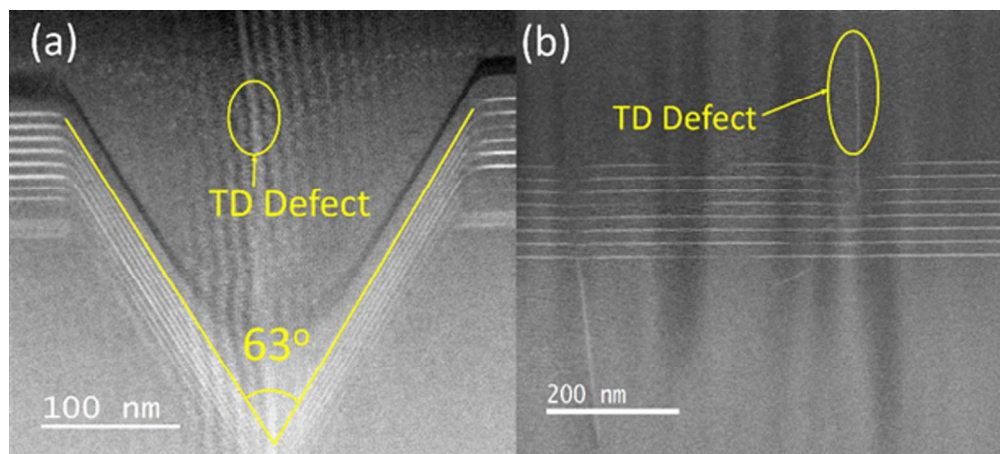


Figure 1 (a) Cross-sectional STEM image of LED1's V-pits with MQW thinning on the $\{101\bar{1}\}$ facets. (b) Cross-sectional STEM image showing LED2 MQWs with TD defects.

83x37mm (300 x 300 DPI)

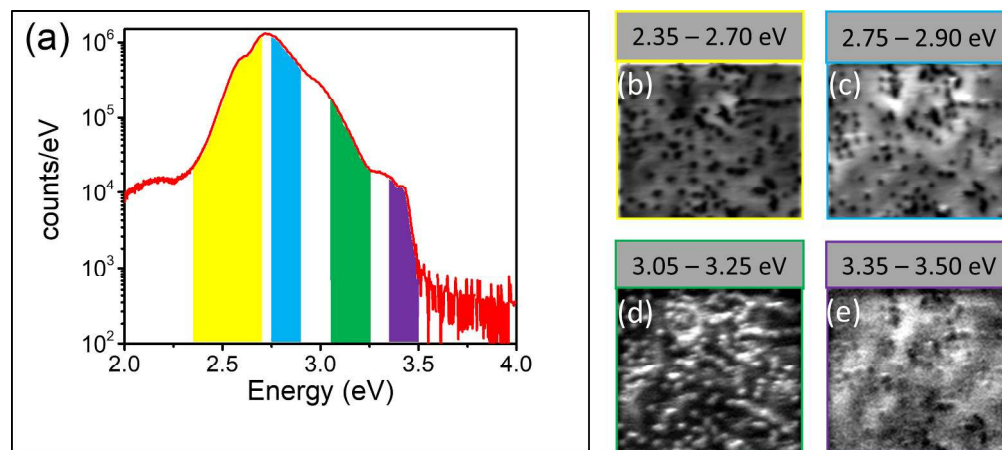


Figure 2 (a) Mean CL emission spectrum of LED1 generated from a $10 \mu\text{m} \times 10 \mu\text{m}$ area hyperspectral map with (b), (c), (d) and (e) CL intensity maps corresponding to the color shaded regions marked on the spectrum.

988x437mm (72 x 72 DPI)

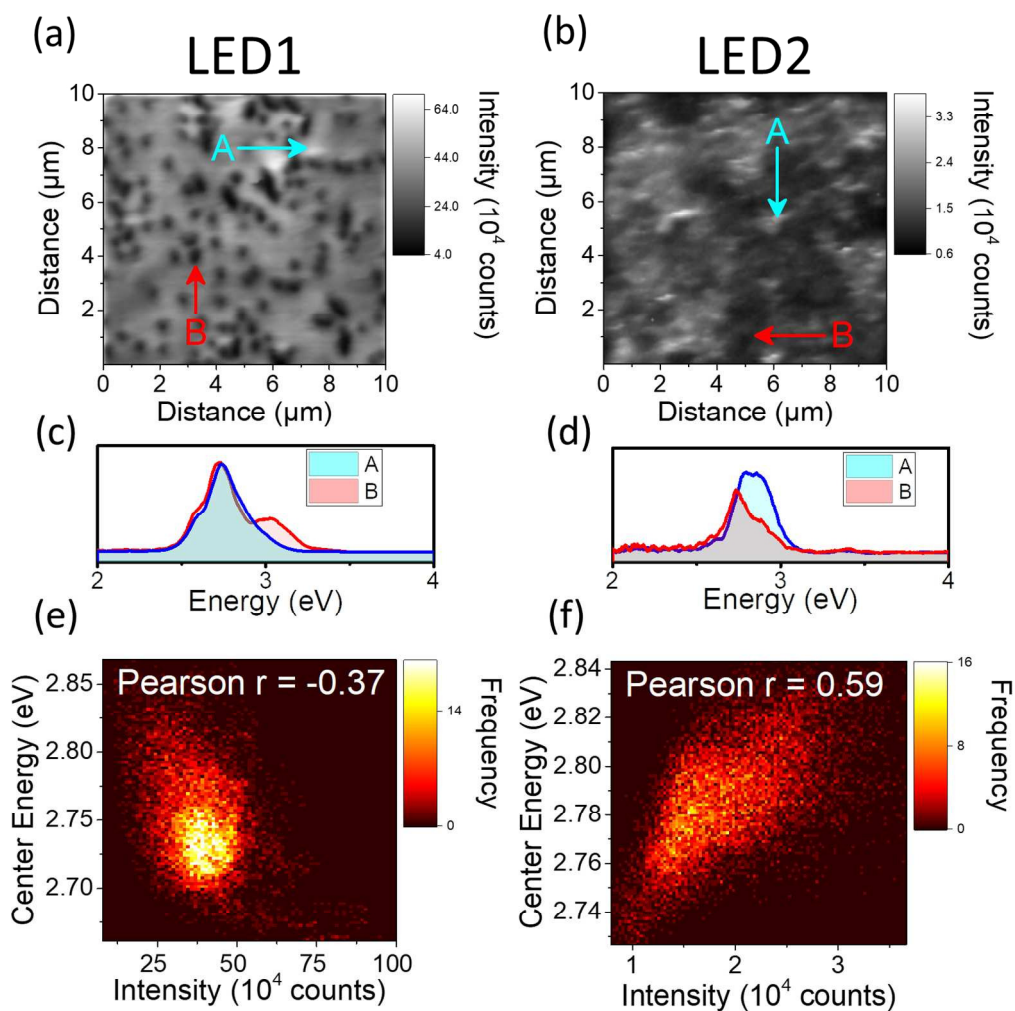


Figure 3 (a) and (b) RT-CL intensity maps of the two LEDs, (c) and (d) CL Spectra of the annotated regions on the micrographs, (e) and (f) Correlation of centroid energy (between 2.4 – 3.2 eV) vs intensity (for LED1 and LED2, respectively).

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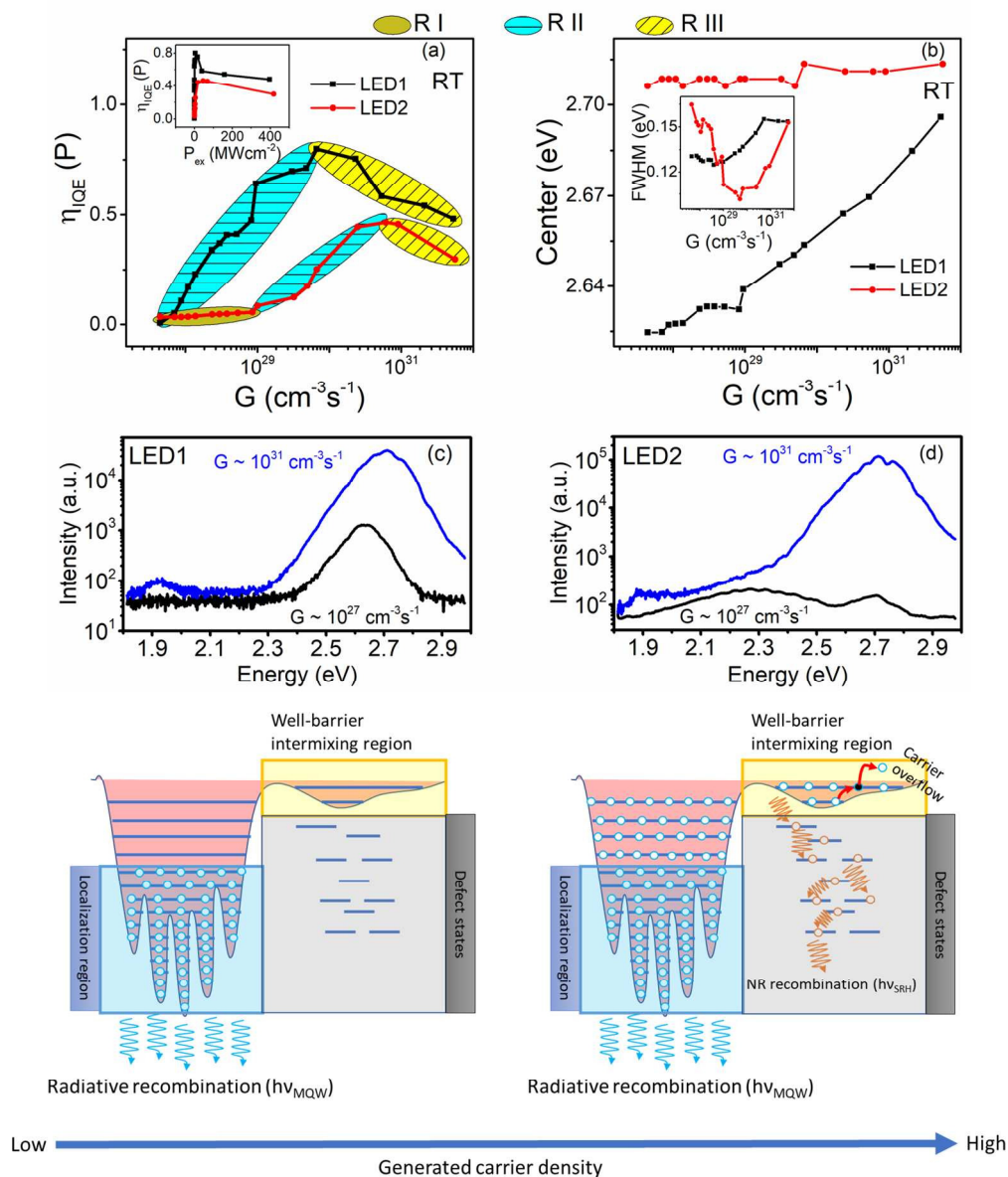


Figure 4 Carrier generation rate dependence of (a) IQE (inset: IQE vs excitation power intensity (linear-scale)) and (b) peak energies (inset: FWHM vs G). PL spectra of (c) LED1 and (d) LED2 at high and low G . (e) the carrier dynamic and droop mechanism of LED1 as G increases.

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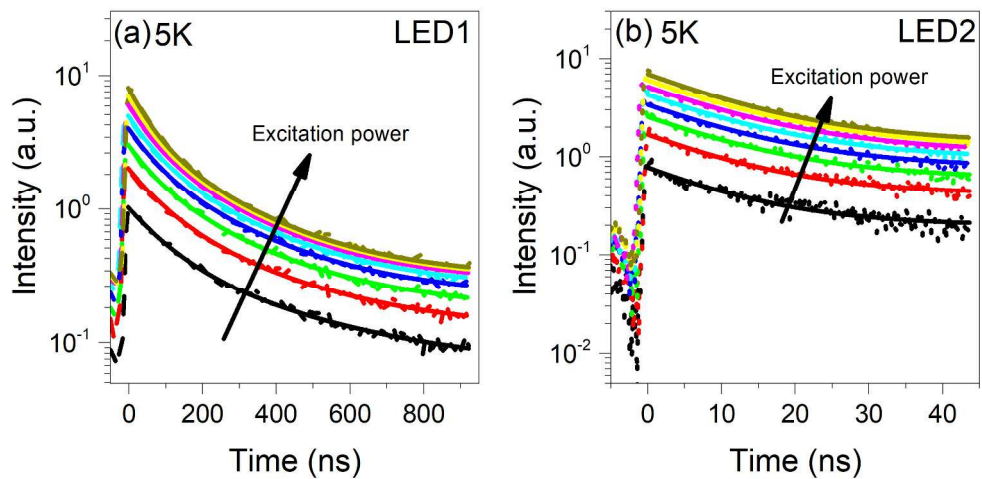


Figure 5 TRPL temporal response at different excitation powers ($\sim 0.044 - 0.76 \text{ MW/cm}^2$) of (a) LED1 and (b) LED2.

247x119mm (300 x 300 DPI)

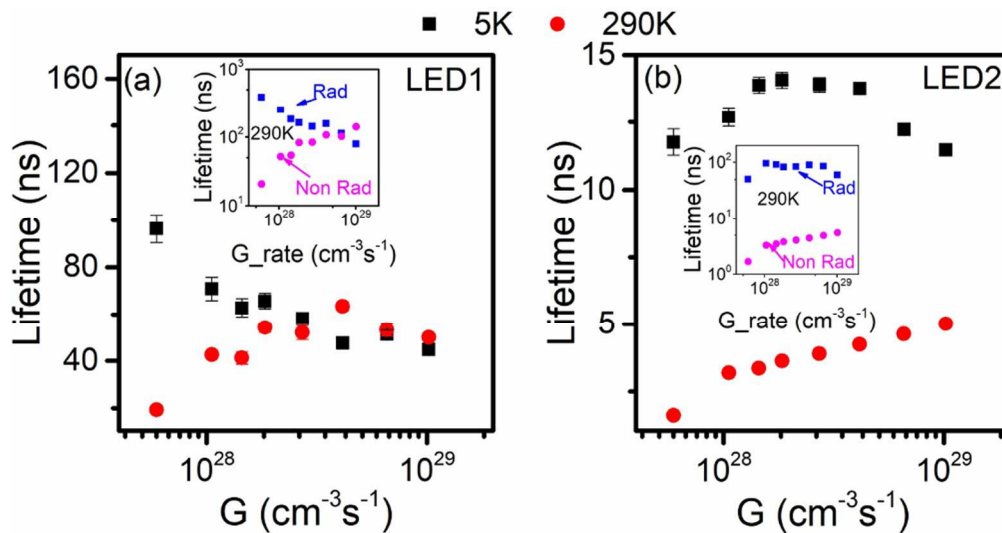


Figure 6 PL lifetimes of (a) LED1, and (b) LED2 as a function of G at 5K (black squares) and 290K (red circles). Insets are the radiative (blue squares) and non-radiative (magenta circles) lifetimes with respect to G at 290K.

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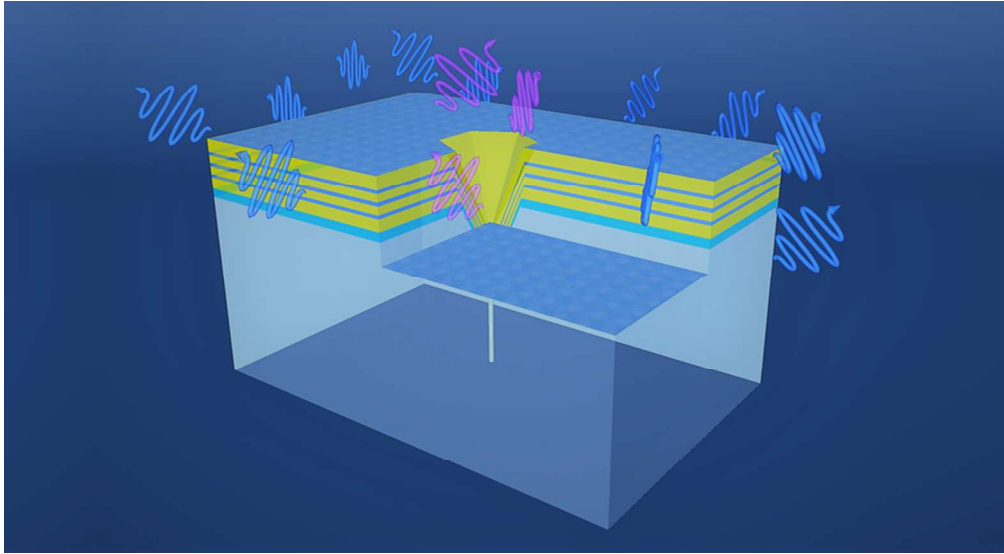


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88x48mm (300 x 300 DPI)