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Temperature and excitation dependent stimulated emission and spontaneous emission in InGaN epilayer

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Temperature and excitation dependence of stimulated emission and spontaneous emission in InGaN epilayer*

Xuee An(安雪娥)[1], Zhengjun Shang(商正君)[1], Chuanhe Ma(马传贺)[1], Xinhe Zheng(郑新和)[2], Cuiling Zhang(张翠玲)[3], Lin Sun(孙琳)[1], Fangyu Yue(越方禹)[1], Bo Li(李波)[1,†], and Ye Chen(陈晔)[1,‡]

[1] *Key Laboratory of Polar Materials and Devices, East China Normal University, Shanghai* 200241, *China*
[2] *Department of Physics, Beijing University of Science and Technology, Beijing* 100083, *China*
[3] *Shanghai Key Laboratory of Green Chemistry and Chemical Processes, Department of Chemistry, School of Chemistry and Molecular Engineering, East China Normal University, Shanghai* 200241, *China*

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Temperature and excitation dependent photoluminescence (PL) of InGaN epilayer grown on *c*-plane GaN/sapphire template by molecular beam epitaxy (MBE) has been systematically investigated. The emission spectra of the sample consisted of strong multiple peaks associated with one stimulated emission (SE) located at 430 nm and two spontaneous emissions (SPE) centered at about 450 nm and 480 nm, indicating the co-existence of shallow and deep localized states. The peak energy of SE exhibiting weak s-shaped variation with increasing temperature revealed the localization effect of excitons. Moreover, an abnormal increase of the SPE intensity with increasing temperature was also observed, which indicated that the carrier transfer between the shallow and deeper localized states exists. Temperature dependent time-resolved PL (TRPL) demonstrated the carrier transfer processes among the localized states. In addition, a slow thermalization of hot carriers was observed in InGaN film by using TRPL and transient differential reflectivity, which is attributed to the phonon bottleneck effect induced by indium aggregation.

Keywords: InGaN, stimulated emission, spontaneous emission, carrier transfer

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1. Introduction

InGaN ternary-alloy semiconductors have attracted considerable attention because of their numerous applications in light emitting diodes and laser diodes, due to their adjustable bandgap from ultraviolet to infrared region.[1–4] However, besides the high-density threading dislocations due to the epitaxial growth on high mismatched sapphire substrates, indium incorporation will lead to severe indium aggregation[5,6] and phase separation.[7,8] Thus, occasional multiple peak (MP) emissions, near band edge excitions, and one or more broad photoluminescence (PL) bands located at lower energies were observed due to the complex band tail states.[9,10] The MP lines had been suggested to be related to the In-rich quantum dots[11] originated from the indium segregation effect and the V-defects initiated by threading dislocations.[12] Although tremendous efforts have been made, to our knowledge, most of the published articles have focused on the carrier thermal population among the SPE luminescent states,[13–15] the emission mechanisms and carrier thermal transfer among the MP lines under the condition of stimulated emission (SE) have left much to be explored. Actually, the emission mechanisms in InGaN film were perplexed due to the existences of several complex processes such as carrier thermalization and carrier transfer in different localization states, enhanced carrier recombination through SE,[16–18] and phonon bottleneck effect[19] caused by the inhomogeneous distribution of indium.

We have reported carrier thermalization under stimulated emission in InGaN epitaxial layer at room temperature.[20] Here, we mainly focused on the studies of the carrier transfer between SE and SPE, and also among different localized states by measuring the temperature dependent PL, and time resolved PL. Stimulated emission at about 430 nm dominated the PL spectrum in the In-rich InGaN film at low temperature. Multiple PL bands appeared at the lower energy side with increasing temperature. Clear carrier transfer processes among these localized luminescent states were observed in the temperature-dependent PL spectra, and the time-resolved PL and transient differential reflectivity measurement demonstrated a slow thermalizaiton of hot carriers and the carrier transfer among these localized states.

2. Samples and experimental techniques

The InGaN sample in this work was grown on GaN layer by plasma assisted molecular beam epitaxy (PA-MBE). A 50 nm thick GaN buffer layer was deposited on the sapphire substrate at a growth temperature of 650 ℃ under intermedi-

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†Corresponding author. E-mail: bli@ee.ecnu.edu.cn
‡Corresponding author. E-mail: ychen@ee.ecnu.edu.cn

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ate Ga-rich condition, and then a 150 nm InGaN epilayer was grown on it. The film was characterized by the high-resolution x-ray diffraction (HR-XRD). The diffraction peaks from the (0002) planes of the InGaN epilayer and GaN buffer were observed, which demonstrated that the InGaN epilayer has the same crystal structure as the GaN buffer. In addition, a weak InN diffraction peak was also observed, which indicated the existence of phase separation. The detailed data can be found in the literature.[20] The appearance of high-In content phase has been reported in InGaN film under Ga-rich condition,[7] and could result in some interesting optical behaviors. The content of indium was estimated to be about 17% based on XRD and PL spectroscopy. For the temperature dependent PL spectra measurements, the sample was mounted on the cold head of a helium gas cycling cryogenerator with temperature adjustable range from 4 K to 300 K. The laser pulse with a repetition rate of 80 MHz from a mode-locked Ti-sapphire oscillator was frequency doubled by a b-barium orate (BBO) crystal, and then a 400 nm pulsed laser with 100 fs pulse-width and 4 μJ single-pulse energy was obtained to be used as the excitation source. The beam spot size focused on the sample was about 30 μm in diameter. A monochromator (iHR 550) with a switchable mirror enabled us to collect the time-integrated and time-resolved PL signals simultaneously. The time-resolved PL (TRPL) signals were recorded by a time-correlated single photon counting (TCSPC) system (PicoHarp300) and a Si avalanche photo-diode (APD) with overall response time about 40 ps.

3. Results and discussion

Figure 1 shows the PL spectra of the InGaN epitaxial layer measured under various excitation densities at low temperature 4 K. It can be seen that a strong and narrow PL peak located at about 430 nm dominates the PL spectra. With increasing excitation density, a weak broad PL band appears at lower energy side from 445 nm to 520 nm. The period envelope structure of the weak PL band is due to the Fabry–Perot interference effect of emission. By fitting with a Gaussian function, we obtain the peak energy and intensity of the main PL peak at 430 nm. Inset (i) gives the excitation density dependence of the PL intensity. It can be clearly seen that the PL intensity exhibits a super-linear dependence on the excitation density, which is a typical feature of stimulated emission. The peak energy keeps nearly constant under various excitation densities. Since the excitation energy (3.1 eV) is lower than the band gap of GaN, we attribute the main peak to the stimulated emission originating from InGaN matrix while the weak PL band at the lower energy side is attributed to the spontaneous emission of band tail states in the InGaN matrix. Furthermore, we take into account the penetration depth ($d = 1/\alpha = 1/(1.5 \times 10^5\ \mathrm{cm}^{-1}) = 6.7 \times 10^{-4}$ cm) of the sample, a excitation density of 5.6 μJ/cm^2 roughly corresponds to a carrier density of 1.7×10^{18} cm^{-3}, which is nearly one order of magnitude higher than that of SPE and is large enough for SE in InGaN sample.[21,22] Especially, the PL spectrum as a function of excitation density near the SE threshold at 300 K is given in inset (ii) of Fig. 1. It shows a clear rising up of the sharp SE peak from the higher energy side of the SPE band. In fact, the sharp peak appears randomly near the threshold of excitation density, and then rises up as a strong main peak with increasing excitation further. It indicates that the SE is related to the localized luminescent centers. Although no cavity was grown in the sample, the material imperfections, such as cracks, burned spot,[23] and localized islands structure due to In aggregation may play the role of optical feedback.[2] These mechanisms could contribute to the stimulation emission in our sample. Such an evolution of turning from SPE to SE is not observed clearly at $T = 5$ K due to the lower threshold of SE at low temperatures.

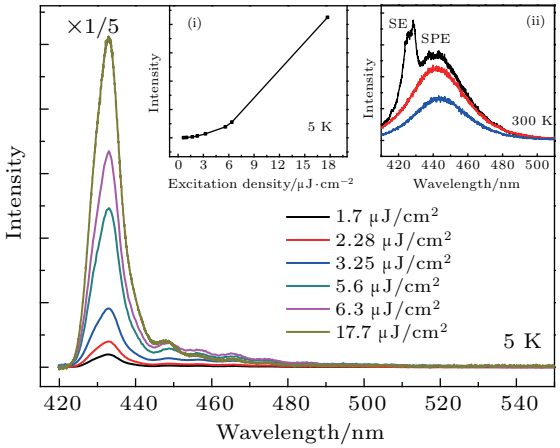


Fig. 1. PL spectra of InGaN film under different excitation densities at low temperature 4 K. Inset (i) shows the PL intensity of SE peak as a function of excitation density. Inset (ii) shows the PL spectra under different excitation densities at 300 K.

Notice that, the SE peak dominates the spectra throughout the whole excitation density range, while the SPE band is rather weak and can only be observed clearly under high excitation density at low temperature (4 K). This can be explained as follows. First, the threshold of SE is lower at low temperature because of the higher luminescence efficiency associated with the localization of the excitons.[16] Large amounts of carriers are consumed in the SE processes instead of transferring to the deeper localized states. Second, the carrier accumulation effect leads to the larger possibility of SE at the higher localized states under excitation condition of 80 MHz high repetition rate.

Figure 2 exhibits the PL curves recorded at different temperatures from 4 K to 300 K for InGaN film under the excitation density of 5.6 μJ/cm^2. At low temperatures (4–40 K),

the SE peak located at 430 nm dominates the spectra. With increasing temperature above 40 K, two broad peaks labeled P_A and P_B, centered at about 450 nm and 480 nm, appear at the lower energy side of the SE band as shown in the inset of Fig. 2. These two broad peaks can be attributed to SPE originating from the dot-like or island-like regions, which have different degrees of localization. Careful analysis shows that P_A dominates the SPE band at low temperatures. However, with increasing temperature to above 150 K, the lower energy peak P_B becomes the dominant SPE emission. This suggests that the thermal population and energy relaxation take place between the shallow and deep localized states.

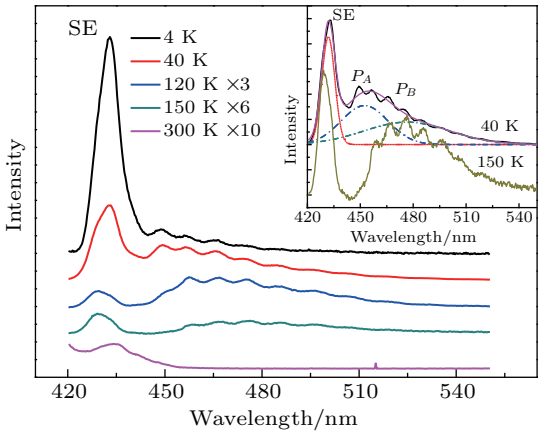


Fig. 2. Temperature-dependent PL spectra under the excitation density of 5.6 μJ/cm^2. The inset shows the typical PL spectra at 40 K and 150 K.

It can be seen that the intensity of SE strongly decreases with increasing temperature, while the intensity of SPE exhibits an abnormal increase at the beginning and then decreases as the temperature further increases. The intensity of SPE decreases slowly compared with that of SE, which makes the SPE to be prominent. With further increasing to room temperature (RT), the SPE band vanishes and the SE dominates the PL spectra again. These temperature dependent PL spectra indicate that the two recombination channels of SE and SPE are relevant to each other and the thermal population of carriers may take place among these emission states.

In an effort to understand carrier transfer processes among these emission states, we have measured the temperature dependent PL spectra under low and high excitation densities (5.6 μJ/cm^2 and 17.7 μJ/cm^2. Figure 3(a) shows the PL peak energies of the SE and SPE bands in InGaN film with increasing temperature from 4 K to 300 K. It can be seen that the peak energy of SE does not follow the typical temperature dependent band gap shrinkage obtained from Varshini's empirical equation $E(T) = E(0) - \alpha T^2/(T + \beta)$, where $E(0)$ is the transition energy at 0 K, and α and β are Varshini thermal coefficients. This shows a weak S-shaped behavior under both low and high excitation densities. The SE peak energy keeps almost unchanged in temperature range from 4 K to 40 K. With increasing temperature to beyond 40 K, the SE peak energy shows a weak blue-shift. As the temperature is further increased to above 150 K, the peak energy of SE becomes consistent with Varshini's equation. In contrary, the peak energies of SPE peaks PA and PB exhibit a monotonous red-shift with increasing temperature. The energy shift of the SPE peaks is about 169 meV, which is much larger than the band gap shrinkage of 36 meV according to the Varshini equation.[23] We attribute the monotonous red-shift of SPE to the deeper localized states compared to that of SE.

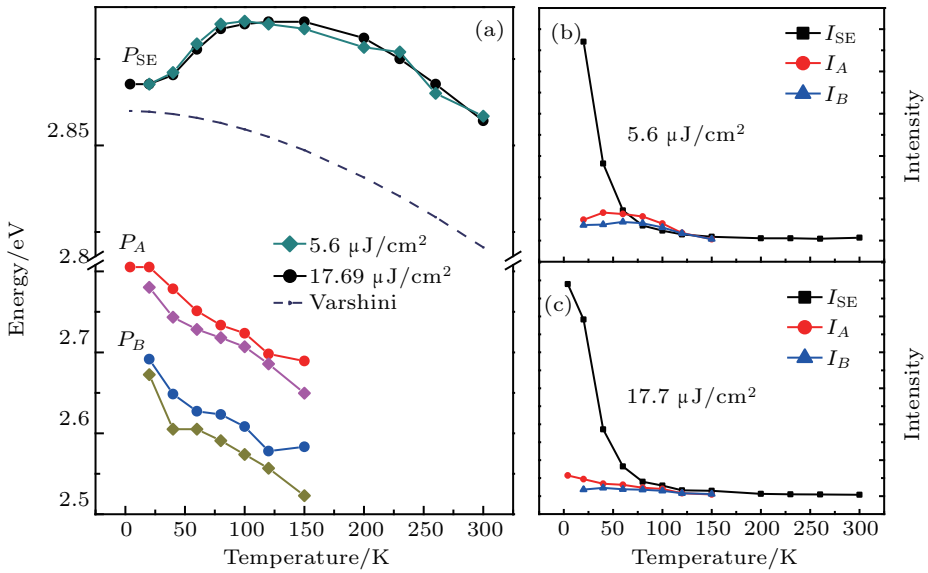


Fig. 3. (a) The peak energies of SE and SPE as a function of temperature. The dot line shows the Varshini bandgap shrinkage of InGaN from Ref. [24]. (b) and (c) PL intensities of the three peaks as a function of temperature under excitation densities of 5.6 μJ/cm^2 and 17.7 μJ/cm^2, respectively.

Figures 3(b) and 3(c) show the PL intensities of the three peaks as a function of temperature under excitation densities of 5.6 μJ/cm^2 and 17.7 μJ/cm^2, respectively. At temperatures from 4 K to 70 K, the PL intensity of SE drops dramatically while the intensity of SPE shows an abnormal increasing evolution under low excitation condition as shown in Fig. 3(b). It indicates that carrier thermal population and energy relaxation take place between the emission states of SE and SPE. It is worthy noting that the SPE intensity does not increase with increasing temperature under high excitation density of 17.7 μJ/cm^2.

From the temperature dependent PL energy and intensity of the SE and SPE peaks, we can see that the anomalous S-shape behavior of the PL peak energy of SE takes place accompanying with a rapid decrease of the PL intensity of SE at low temperature. Similar anomalous S-shape shift of the PL energy has been observed by several groups[24–27] and was attributed to the existence of localized states caused by imperfections in the InGaN layer. It is worthy to note that the SE peak energy keeps almost unchanged at temperatures from 4 K to 40 K. This is quite different from the previous reports[20] that the typical S-shaped behavior of exciton SPE exhibits a decreasing trend at low temperature. This happens because the carriers are randomly distributed in the localized potential minima at low temperature. As the temperature increases from 4 K to 40 K, weakly localized carriers will be thermally activated and relax down into the lowest potential via hopping.[10] It should be noted that the reported anomalous S-shape behavior was observed in spontaneous emission of exciton, while in our case the main PL peak is stimulated emission. The mode selection of SE and larger localized energy in our sample may lead to some pinning effect of the SE peak. Thus, an almost unchanged SE is observed at low temperatures below 40 K.

As the temperature increases from 40 K to 150 K, both PL intensities of the SE and SPE peaks exhibit a continuous decreasing, which indicates the onset of efficient losses caused by non-radiative recombination (NRR) of some delocalized carriers.[14] More important phenomenon is that increasing temperature (above 40 K) enables activate carriers to occupy higher-energy levels of the localized states, thus resulting in the blue-shift of the peak energy as large as 33 meV up to $T \approx 100$ K. Accordingly, the PL intensity of the SE peak drops dramatically, while the intensities of the SPE peaks show an abnormal increasing evolution under low excitation condition (5.6 μJ/cm^2) as shown in Fig. 3(b). The SPE component becomes prominent from 70 K to 150 K under low excitation condition. This indicates that the carrier transfer from SE states to SPE states occurs with increasing temperature even under the condition of stimulated emission. It is worth noting that the raising tendency of SPE intensity with increasing temperature is not observed under the high excitation density of 17.7 μJ/cm^2 as shown in Fig. 3(c). This can be interpreted by the saturation of emission states of SPE under high excitation density. The lower energy localized SPE states have been filled up under high excitation density, thus there are no enough empty states to accept the thermal populated carriers from the higher energy SE states.

It should be noted that thermal population and carrier transfer occur not only between the SE and SPE states, but also between the shallow and deep SPE states. With increasing temperature, the main PL intensity of SPE transfers from P_A to P_B. Moreover, the red-shift of SPE is faster than the shrinkage of bandgap of InGaN. These results can be understood in the framework of thermal population and carrier transfer among the localized SPE states. This means that the localized state rather than the composition is the dominant pathway to make the photons in the InGaN luminescent layer.[13]

With a further increase to room temperature, the SPE component vanishes and the PL spectrum is dominated by the SE component. Considering that majority photo-generated carriers are captured by high-density NRR centers, it is not difficult to understand that the SPE vanishes under high temperature. Moreover, the peak evolution of SE as a function of temperature is in accordance with that of Varshini function in high temperature range, which indicates that the band shrinkage is related to the thermal activation rather than the localization effect.[15] Above 150 K, the photo-generated carriers have enough energy to escape out from the deep localized centers and the localization effect can be neglected.

To explore the ultrafast carrier dynamics in InGaN film in details, temperature-dependent TRPL of the InGaN film has been measured by TCSPC technique under the excitation density of 17.7 μJ/cm^2. Figure 4(a) exhibits the four typical normalized temporal evolutions of the SE intensity at different temperatures. For comparison, the temporal evolution of the SPE intensity at 4 K is also plotted. We can see that all the TRPL curves measured at 430 nm at different temperatures show a slow rise process and a mono-exponential decay. The instrument response function (IRF) is also given for comparison. Thus, the relative slow rise process is not due to the system response. Moreover, the TRPL curves of the SE peak at 430 nm exhibit similar rise and decay processes to that of the SPE band at 450 nm, except of a little faster decay process. According to the previous publications, the lifetime of SE in InGaN is in the order of ps.[28] The lifetime and rise time of the SE process might be considered to be significantly shorter than the overall system response time (40 ps). Therefore, the PL decay curves is actually the component of spontaneous emission.[20]

All of the decay curves exhibit mono-exponential rise and decay processes, and an exponential decay function $I = I\exp(-t/\tau)$ convoluted with the IRF is used to fit the curves

to obtain the lifetime. The PL lifetime for both emissions decreases with increasing temperature, especially at a temperature about $T = 40$ K. At a temperature about 40 K, the delocalization induced by thermal activation and carrier transfer takes place, thus leading to the decrease of lifetime, which is consistent with the anomalous S-shape behavior of the PL peak above. It indicates that the thermal population takes place at the temperature about 40 K.

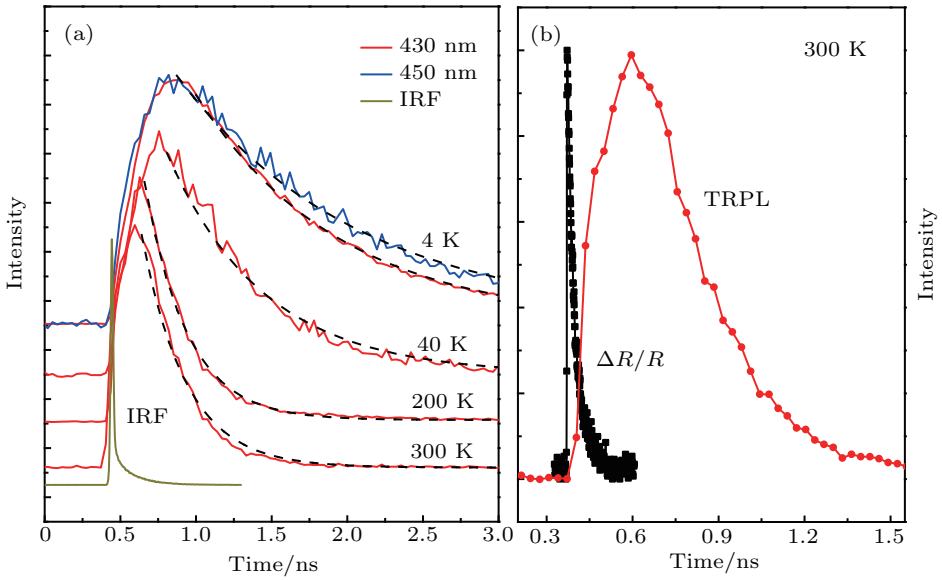


Fig. 4. (a) Time-resolved PL of InGaN film measured by TCSPC at different temperatures. The IRF curve is also plotted. The dashed line represents the lifetime and the dash line represents the rise time of SE. (b) The transient differential reflectivity $\Delta R/R$ and TRPL of InGaN film measured at 300 K.

It is of note that the rise time of PL emission at 430 nm decreases from 429 ps to 124 ps with increasing temperature from 4 K to 300 K. In general, the rise time reflects the thermalization process of photo-generated carriers from the excited states to the band tail states. Thus, our results indicate that the thermalization of carriers is much slower for our InGaN sample. To investigate the ultra-fast carrier dynamics, we have also measured the transient differential reflectivity $\Delta R/R$ of the InGaN film. Figure 4(b) exhibits the comparison of transient $\Delta R/R$ and TRPL at 300 K. It can be seen that transient $\Delta R/R$ shows an instantaneous rising followed by a double-exponential function relaxation process. More importantly, the decay process of $\Delta R/R$ corresponds well to the rising process of the TRPL curves. In general, the rising of $\Delta R/R$ corresponds to the excitation of carriers from ground to excited states, and the decay process is related to the thermalization and accumulation of carriers from the excited states to the localized tail states. It should be noted that a 400 nm pulse was used to pump the sample, while an 800 nm pulse was used to probe the $\Delta R/R$ in our measurements. Therefore, the double-exponential decay processes obviously correspond to the subsequent multi-stage decay from the excited states to the localized tail states. As we discussed above, the stimulated emission comes from the localized luminescent centers. Consequently, the photo-generated carriers first relax to the bottom of the conduction band rapidly by migration, and they then relax down into the deeper localized states subsequently by tunneling. The potential depth depends on the concentration of In, which changes with the change of special position. Such a slow cooling process of hundreds of picoseconds is related to the phonon bottleneck effect induced by the phase separation and indium aggregation in InGaN. A detailed analysis and discussion of this point has been given in our previous published paper.[20]

4. Summary

In conclusion, we have investigated the time integrated PL and TRPL spectra of InGaN epitaxial layer over the temperature range from 4 K to 300 K. We observe obvious multiple peaks emissions at 430 nm, 450 nm, and 480 nm, which are considered to be associated with SE, SPE from shallower and deeper localized states, respectively. The peak energy of SE exhibits a weak S-shaped behavior with the increase of temperature, indicating the localization and thermal activation of the excitions. The abnormal increase of the SPE intensity with increasing temperature is clearly seen, and can be explained by the carrier transfer from the localized SE states to SPE states, and from shallow to deep localized states via tunneling. This carrier transfer process among these localized luminescent states is also demonstrated in the temperature dependent TRPL. These observations are expected to reveal a distinct picture of dynamic processes of photo-generated carriers in InGaN.

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