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Method for determining the radiative efficiency of GaInN quantum wells based on the width of efficiency-versus-carrier-concentration curve

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We report a method to determine the radiative efficiency (RE) of a semiconductor by using room-temperature excitation-dependent photoluminescence measurements. Using the ABC model for describing the recombination of carriers, we show that the theoretical width of the RE-versus-carrier-concentration (n) curve is related to the peak RE. Since the normalized external quantum efficiency, $EQE_{normalized}$, is proportional to the RE, and the square root of the light-output power, $\sqrt{LOP}$, is proportional to n, the experimentally determined width of the $EQE_{normalized}$-versus-n curve can be used to determine the RE. We demonstrate a peak RE of 91% for a $Ga_{0.85}In_{0.15}N$ quantum well. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4770317]

Recently, III-V nitride-based light-emitting diodes (LEDs) have received much interest because of their demonstrated ability to generate high-quality white light with a much higher efficiency than other lighting technologies.[1,2] GaInN LEDs are highly efficient despite a high threading dislocation density of 10^7–10^9 cm^{-2},[3,4] which is fundamentally different from the III-V arsenide and III-V phosphide material systems. To understand the high efficiency of the III-V nitride material system, several research groups investigated the radiative efficiency (RE) in GaInN/GaN quantum well (QW) structures.[5–7] The RE is defined as the fraction of carriers recombining radiatively in the active region. The radiative efficiency is related to the internal quantum efficiency (IQE) and the injection efficiency (IE) by IQE = RE × IE, where the IQE is defined as the ratio of the number of photons emitted from the active region to the number of electrons injected into the device. The distinction between RE and IE allows one to identify the factor limiting the IQE. Finally, the light-extraction efficiency (LEE) relates the external quantum efficiency (EQE) to the IQE, that is, EQE = IQE × LEE.

One method for determining RE is temperature-dependent photoluminescence (PL) measurements conducted over a range of excitation density. By assuming 100% RE at cryogenic temperatures, e.g., 4 K, the RE at 300 K can be determined from the ratio between the efficiency at 300 K and cryogenic temperature. Another method for determining RE is time-resolved PL (TRPL) measurements conducted at different excitation densities or temperatures.[8,9] In TRPL, a short pulse laser is absorbed by an active material and the time dependence of photoluminescence decay is measured. This method is useful to understand the temporal response of carrier recombination and transport in the active material. There is yet another method, based on electroluminescence measurements, which can measure the RE under LED operating conditions.[5,10] Using this method, Ryu *et al.* found a RE over 85% in GaInN QW samples measured at 300 K.[5]

In this paper, we present a method that allows one to determine the RE of a semiconductor QW structure based on resonant-excitation PL measurements that are carried out over a wide range of excitation powers. Based on the ABC model, we derive a mathematical relationship between peak radiative efficiency (RE_{peak}) and the width of the experimental $EQE_{normalized}$-versus-n curve. Using this method, we analyze a GaInN QW structure and determine its RE_{peak}.

The ABC model is well-known for its effectiveness to describe carrier recombination in QW structures; however, the applicability of the ABC model is limited to cases where all carriers are electrically injected into the QWs (or optically generated inside the QWs) and remain in the QWs until they recombine (i.e., carriers do not escape). The ABC model describes three recombination mechanisms in an active material: Shockley-Read-Hall (SRH), radiative, and Auger recombination.[11] In the ABC model, the total recombination rate R is given by

$$R(n) = A_{SRH}n + Bn^2 + C_{Auger}n^3, \quad (1)$$

where A_{SRH}, B, C_{Auger}, and n is the SRH, radiative, Auger coefficient, and the carrier concentration, respectively. Based on the ABC model, the RE of an active material can be expressed as

$$RE(n) = Bn^2/R(n) = Bn^2/(A_{SRH}n + Bn^2 + C_{Auger}n^3). \quad (2)$$

Based on this equation, we can plot theoretical RE-versus-n curves for different values of A_{SRH}, B, and C_{Auger}. These plots illustrate how the RE-versus-n curves depend on the A_{SRH}, B, and C_{Auger} coefficients. For example, changing A_{SRH} from 10^5 to 10^8 s^{-1} (while keeping $B = 10^{-10}$ cm^3/s and $C_{Auger} = 10^{-30}$ cm^6/s constant) illustrates the effect of A_{SRH} on the radiative efficiency value and the shape of the RE-versus-n curve. Such an A_{SRH}-varying series is plotted in

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green color in Figure 1(a). Inspection of the figure reveals that the $\mathrm{RE}_{\mathrm{peak}}$ increases when A_{SRH} decreases. Furthermore, the full-width at half-maximum (FWHM) of the RE-versus-n curve also increases. This suggests that the value of $\mathrm{RE}_{\mathrm{peak}}$ is related to the width of the RE-versus-n curve. Similarly, a C_{Auger}-varying series, plotted in red color in Figure 1(a), shows the same tendency: A curve with a higher $\mathrm{RE}_{\mathrm{peak}}$ results in wider RE-versus-n curve. This trend is maintained even when changing the radiative coefficient B, as shown in Figure 1(b). This suggests that there is a fundamental relationship between the $\mathrm{RE}_{\mathrm{peak}}$ of a semiconductor and the width of RE-versus-n curve.

The relation between the $\mathrm{RE}_{\mathrm{peak}}$ and the width of RE-versus-n curve can be derived as follows: First, from the condition that the first derivative of RE with respect to n equals zero, i.e., $\mathrm{dRE}/\mathrm{d}n = 0$, the peak-efficiency carrier concentration, $n_{\text{RE-peak}}$, can be determined

$$n_{\text{RE-peak}} = \sqrt{A_{\mathrm{SRH}}/C_{\mathrm{Auger}}}. \tag{3}$$

Inserting this concentration into Eq. (2) yields $\mathrm{RE}_{\mathrm{peak}}$

$$\begin{aligned}\mathrm{RE}_{\mathrm{peak}} &= \mathrm{RE}(n = n_{\text{RE-peak}}) = \\ &= \frac{B n_{\text{RE-peak}}^2}{A_{\mathrm{SRH}} n_{\text{RE-peak}} + B n_{\text{RE-peak}}^2 + C_{\mathrm{Auger}} n_{\text{RE-peak}}^3} \\ &= \frac{1}{1 + 2\sqrt{A_{\mathrm{SRH}} C_{\mathrm{Auger}}}/B}.\end{aligned} \tag{4}$$

Second, we employ the even symmetry of the RE-versus-n curve under the ABC model.[12] That is, for any radiative efficiency value, i.e., $\alpha\, \mathrm{RE}_{\mathrm{peak}}$ $(0 < \alpha < 1)$, there are two carrier concentrations n_1 and n_2 located symmetrically, i.e., at equal distance from $n_{\text{RE-peak}}$ $(n_1 < n_{\text{RE-peak}} < n_2)$ having the same radiative efficiency. That is,

$$\mathrm{RE}(n_1) = \mathrm{RE}(n_2) = \alpha\, \mathrm{RE}_{\mathrm{peak}}. \tag{5}$$

By using Eq. (2) to express $\mathrm{RE}(n_1)$ and $\mathrm{RE}(n_2)$ and using that $\mathrm{RE}(n_1) = \mathrm{RE}(n_2)$, one obtains

$$n_1 n_2 = A_{\mathrm{SRH}}/C_{\mathrm{Auger}} = n_{\text{RE-peak}}^2. \tag{6}$$

The "distance" between n_1 and $n_{\text{RE-peak}}$, on a logarithmic scale, is $\log(n_{\text{RE-peak}}) - \log(n_1) = \log(n_{\text{RE-peak}}/n_1)$; in the same way, the distance between n_2 and $n_{\text{RE-peak}}$ is $\log(n_2) - \log(n_{\text{RE-peak}}) = \log(n_2/n_{\text{RE-peak}})$. These two distances are equal as can be proven by Eq. (6). Therefore, the width of RE-versus-n curve at radiative efficiency $\alpha\, \mathrm{RE}_{\mathrm{peak}}$ is $\log(n_2/n_1) = 2\log(n_{\text{RE-peak}}/n_1) = 2\log(n_2/n_{\text{RE-peak}})$. The even-symmetry curve of ABC model is illustrated in Figure 1(c).

Finally, we determine the relation between $\mathrm{RE}_{\mathrm{peak}}$ and $\log(n_{\text{RE-peak}}/n_1)$, i.e., the relation between $\mathrm{RE}_{\mathrm{peak}}$ and the width of efficiency curve. By inserting $n = n_2$ into the Eq. (2), and using the relation $n_2 = n_{\text{RE-peak}}^2/n_1$ (see Eq. (6)), we obtain

$$\mathrm{RE}(n = n_2 = n_{\text{RE-peak}}^2/n_1) = \frac{B(n_{\text{RE-peak}}^2/n_1)^2}{A_{\mathrm{SRH}}(n_{\text{RE-peak}}^2/n_1) + B(n_{\text{RE-peak}}^2/n_1)^2 + C_{\mathrm{Auger}}(n_{\text{RE-peak}}^2/n_1)^3} = \alpha\, \mathrm{RE}_{\mathrm{peak}}. \tag{7}$$

Next, we substitute $\sqrt{A_{\mathrm{SRH}}/C_{\mathrm{Auger}}}\; n_{\text{RE-peak}}/n_1$ for $n_{\text{RE-peak}}^2/n_1$. Rewriting Eq. (7) yields

$$\frac{(n_{\text{RE-peak}}/n_1)}{(n_{\text{RE-peak}}/n_1) + (\sqrt{A_{\mathrm{SRH}} C_{\mathrm{Auger}}}/B)\left(1 + (n_{\text{RE-peak}}/n_1)^2\right)} = \alpha\, \mathrm{RE}_{\mathrm{peak}}. \tag{8}$$

Eliminating $\sqrt{A_{\mathrm{SRH}} C_{\mathrm{Auger}}}/B$ by using Eq. (4) yields

$$\left(\frac{n_{\text{RE-peak}}}{n_1}\right)^2 - \left[\frac{2(1 - \alpha\, \mathrm{RE}_{\mathrm{peak}})}{\alpha(1 - \mathrm{RE}_{\mathrm{peak}})}\right]\left(\frac{n_{\text{RE-peak}}}{n_1}\right) + 1 = 0. \tag{9}$$

In this quadratic equation, coefficients A_{SRH}, B, and C_{Auger} do not appear. The equation can be solved analytically. It has two solutions, only one of them physically meaningful

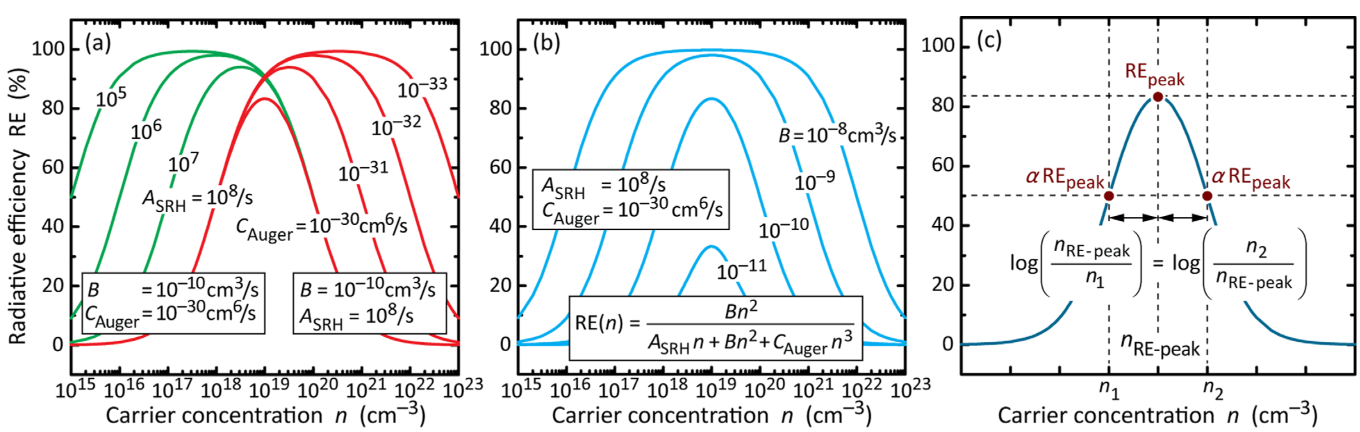

FIG. 1. Calculated RE as a function of carrier concentration n for different values of (a) the A_{SRH} and C_{Auger} coefficient and (b) the B coefficient; (c) the even-symmetry property of the RE-versus-n curve based on the ABC model. Any two carrier concentrations with the same "distance" from $n_{\text{RE-peak}}$ are associated with the same radiative efficiency, i.e., $\alpha\, \mathrm{RE}_{\mathrm{peak}}$.

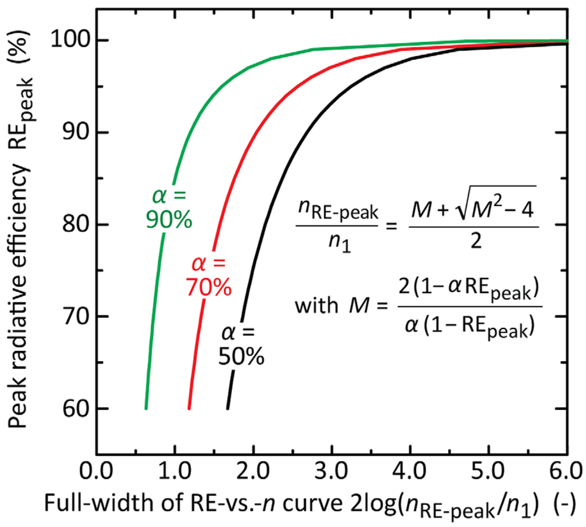

FIG. 2. Calculated relation between RE_{peak} and full-width of RE-versus-n curve at $\alpha = 50\%$ (FWHM), 70%, and 90%.

$$\frac{n_{RE\text{-}peak}}{n_1} = \frac{M+\sqrt{M^2-4}}{2}, \quad \text{where} \quad M = \frac{2(1-\alpha RE_{peak})}{\alpha(1-RE_{peak})}. \tag{10}$$

Therefore, a relation between ($n_{RE\text{-}peak}/n_1$) and RE_{peak} is derived. This relationship between RE_{peak} and the width at efficiency α RE_{peak} is shown in Figure 2 where α is a freely selected parameter at which the width of the *RE*-versus-n curve is considered. For example, when $\alpha = 50\%$, then the width of the RE-versus-n curve is the full-width at half-maximum, i.e., 2 log ($n_{RE\text{-}peak}/n_1$).

To use this method for evaluating the experimental RE of an active region, including its RE_{peak}, we plot the experimentally determined $EQE_{normalized}$-versus-$\sqrt{LOP}$ curve, where $EQE_{normalized}$ is the normalized external quantum efficiency, and LOP is the light-output power. Because $EQE_{normalized}$ can be assumed to be proportional to RE and $\sqrt{LOP}$ can be assumed to be proportional to n (LOP $\propto Bn^2 \propto n^2$ and therefore $\sqrt{LOP} \propto n$), the width of the $EQE_{normalized}$-versus-$\sqrt{LOP}$ curve and the width of the RE-versus-n curve are the same. Note that $\sqrt{LOP}$ equals the mathematical product of a proportionality factor and n. The proportionality factor causes only horizontal shift of the curve along the log n abscissa without changing the shape or width of the RE-versus-n curve. Further, the measured $EQE_{normalized}$ is proportional to the IQE. Consider a 405 nm-laser resonantly exciting a blue-emitting GaInN single QW (SQW) sandwiched by two GaN barriers with flat conduction and valence bands; assume further a conduction-to-valence band offset ratio of 50:50.[13] Under these conditions, the confining barrier heights of the SQW for both electrons and holes is at least 6 kT (k is Boltzmann constant and $T = 300$ K); this makes carrier leakage highly unlikely and IE = 1.0. For experimental conditions where the Auger coefficient is exceedingly small or where the 405 nm laser power is limited, the high-excitation, decreasing branch of the $EQE_{normalized}$-versus-n curve may be difficult to measure. Under these conditions, our method can still be used by using the even symmetry of the $EQE_{normalized}$-versus-n curve. That is, if the full width is not revealed in experiments, the half width can be used to determine the RE_{peak} value. In theory, the half width of the $EQE_{normalized}$-versus-n curve can be measured at a normalized efficiency of, e.g., $\alpha = 50\%$, to determine RE_{peak}. In practice, we are limited in our choice of α since, for high-quality samples, the measured efficiency may not decrease to such a low value, i.e., 50%. Therefore, we choose an α as low as we can, so that the half width is as wide as possible; this minimizes the error incurred when determining the RE_{peak}.

However, when there is a significant built-in potential inside the sample (such as a built-in potential caused by a pn junction), carrier leakage can occur, particularly at low excitation densities where the built-in potential is strong. In this case, the injection efficiency no longer can be assumed to be 100% and the width of the RE-versus-n curve is distorted because of the smaller injection efficiency.

To investigate the determination of RE using the method described above, we prepared two different GaInN/GaN SQW samples, both grown by metal-organic vapor-phase epitaxy. One sample is an n-type-GaN/GaInN-SQW/n-type-GaN epitaxial structure (sample DNZ-2762), whereas the other sample is an n-type-GaN/GaInN-SQW/p-type-GaN epitaxial structure (sample DNZ-2764). As a further detail, we note that each SQW sample has a ~200 nm-thick n-type $In_{0.02}Ga_{0.98}N$ underlayer between the n-type GaN and the SQW. The only difference in the sample structures is the doping in the top layer. Sample DNZ-2762 whose SQW is sandwiched between two equally doped n-type GaN layers has no built-in pn-junction potential. Because of the lack of a pn-junction potential, a better confinement of carriers in the SQW (and no carrier leakage) can be expected. Excitation-dependent PL measurements at 300 K are conducted on these two samples. The excitation source is a 405 nm semiconductor laser with optical power up to 300 mW. The laser pulse duration is 10 μs with a 1% duty cycle to avoid heating effects. The laser is focused onto the sample and the average radius of the circular laser spot is 13 μm. PL is collected by an optical fiber and is analyzed by a spectrometer.

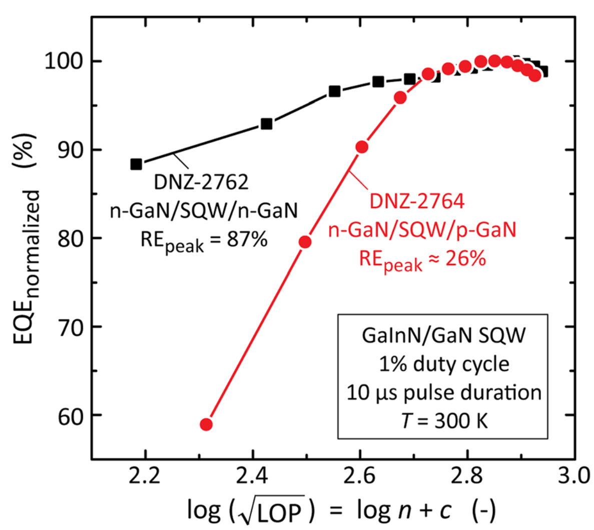

FIG. 3. The $EQE_{normalized}$-versus-$\sqrt{LOP}$ of the n-GaN/SQW/n-GaN and n-GaN/SQW/p-GaN samples and IQE_{peak} calculated from the half-width at $T = 300$ K (c is a constant that indicates an abscissa shift).

The two samples have the same peak emission wavelength of 437 nm. Based on the spectra, the PL intensity is calculated by integrating the spectral intensity over wavelength. Accordingly, the relative EQE and $EQE_{normalized}$ can be evaluated at each excitation power density. The $EQE_{normalized}$-versus-$\sqrt{LOP}$ curves of the two samples are shown in Figure 3. The sample without built-in potential has a higher efficiency and shows wider half-width than the sample with built-in potential. The measured half-width of $EQE_{normalized}$-versus-$\sqrt{LOP}$ curve corresponds to RE_{peak} of 87% for the sample without built-in potential. This value indicates a high sample quality. Given that the sample was grown under standard growth conditions, the result also suggests that the RE_{peak} of highly optimized GaInN MQW structures could well exceed 90%. Using the method described in this paper, we obtain a RE_{peak} of 26% for the sample with built-in potential. Since the two samples are grown under the same growth conditions for the n-type GaN and the SQW, the crystal quality of the active region can be assumed to be the same, that is, the SRH coefficient A_{SRH} and radiative coefficient B can be assumed the same. Therefore, the lower RE_{peak} in the p-SQW-n sample can be attributed to carrier leakage. We suggest that carrier leakage in the p-SQW-n sample results from the built-in potential. We note parenthetically that for the p-SQW-n the measured RE_{peak} may better be denoted as IQE_{peak}, since carrier leakage cannot be excluded. As for the sample without built-in potential, carrier leakage is minimized or eliminated because of good carrier confinement and flatter bands on both sides of SQW of the n-SQW-n sample.

To independently verify the method presented here, we conducted temperature-dependent photoluminescence experiments on the n-type-GaN/SQW/n-type-GaN sample. The excitation laser in the photoluminescence setup is a Kr-ion laser operating at 1% duty cycle with 10 μs pulse duration, and the sample, mounted in a cryostat, was measured at liquid He temperature ($T = 4$ K) and 300 K. The efficiency-versus-excitation-power curves at 4 K and 300 K are shown in Figure 4(a). Assuming that the RE_{peak} at 4 K is 100%, the RE_{peak} at 300 K is determined from the 300 K to 4 K integrated-intensity ratio and found to be 91%. As a next step, we convert the measured data shown in Figure 4(a) to an $EQE_{normalized}$-versus-$\sqrt{LOP}$ curve, shown in Figure 4(b), so that the width of this curve can be directly read from the figure (the width will be used to determine the RE_{peak}). In Figure 4(b), we have shifted the curves along the abscissa to align the peak–efficiency concentration to log $n = 0.0$. The wider width of the 4 K data shows that the sample at 4 K has a higher efficiency than at 300 K, as expected. Using Figure 4(b), we choose a pair of points, i.e., the peak efficiency point and another point with lower efficiency, measure the horizontal separation between these two points and then determine the RE_{peak} based on Eq. (10). We choose pairs of points whose horizontal separation is greater than 0.5 to have wide width (and thus a small uncertainty). In Figure 4(c), the RE_{peak} determined from the width of the $EQE_{normalized}$-versus-$\sqrt{LOP}$ curve shows similar values, independent of the α that we choose. Using this procedure, the sample is determined to have a RE_{peak} of 99% at 4 K, while it has a RE_{peak} of 91% at 300 K (average value). Therefore, the method presented here, based on the width of the $EQE_{normalized}$-versus-n curve, agrees with the results inferred from temperature-dependent PL measurements.

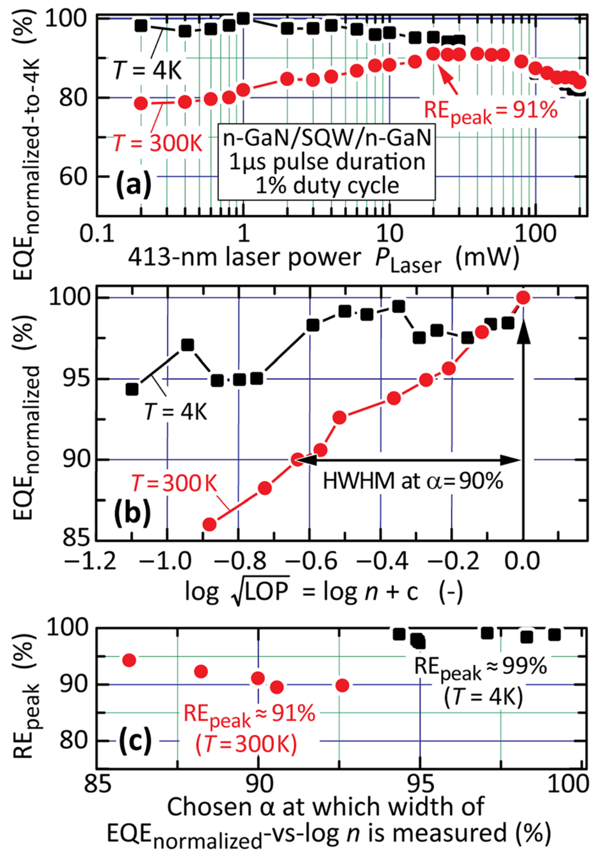

FIG. 4. (a) Relative-efficiency-versus-excitation-laser-power at 4 K and 300 K for the n-type-GaN/SQW/n-type-GaN sample; the efficiency is normalized to the peak efficiency at 4 K. By assuming 100% efficiency at 4 K, one obtains a RE_{peak} of 91% at 300 K. (b) $EQE_{normalized}$-versus-$\sqrt{LOP}$ obtained from the data shown in (a). The half-width can be measured and used to determine RE_{peak}. (c) RE_{peak} determined from the half-width shown in (b). At $T = 4$ K, the sample shows a RE_{peak} of 99% while at $T = 300$ K it shows a RE_{peak} of 91%, in agreement with the temperature-dependent PL results.

In conclusion, we introduce a quantitative method, based on the ABC model, to determine RE_{peak} from the width of the RE-versus-n curve or the experimentally determined $EQE_{normalized}$-versus-$\sqrt{LOP}$ curve as measured by resonant optical excitation photoluminescence measurements on an active material, e.g., a semiconductor QW structure. Using this method, we demonstrate a peak RE of 87%–91% for a GaInN QW emitting at 437 nm.

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