Analysis of Interdiffused InGaN Quantum Wells for Visible Light-Emitting Diodes

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Abstract—Interdiffused InGaN quantum wells (QWs) with various interdiffusion lengths \(L_d\) are comprehensively studied as the improved active region for Light-Emitting Diodes (LEDs) emitting in the blue and green spectral regime. The electron-hole wavefunction overlap \(\Gamma_{e,h}^{c}\), spontaneous emission spectra, and spontaneous emission radiative recombination rate \(R_{sp}\) for the interdiffused InGaN QWs are calculated and compared to that of the conventional InGaN QWs emitting in the similar wavelengths. The calculations of band structure, confined energy levels, electron and hole wavefunctions, and spontaneous emission radiative recombination rate \(R_{sp}\) are based on the self-consistent \(6\)-band \(k\cdot p\) method, taking into account the valence band mixing, strain effect, spontaneous and piezoelectric polarizations and carrier screening effect. Studies indicate a significant enhancement of the electron-hole wavefunction overlap \(\Gamma_{e,h}^{c}\) and the spontaneous emission radiative recombination rate \(R_{sp}\) for the interdiffused InGaN QWs. The improved performance for the interdiffused InGaN QWs is due to the modification of the band lineups at the InGaN-GaN interfaces, which leads to the enhancement of the electron-hole wavefunction overlap significantly.

Index Terms—InGaN quantum wells (QWs), light-emitting diodes (LEDs), interdiffusion.

I. INTRODUCTION

-NITRIDE semiconductor based quantum wells (QWs) as active region for light-emitting-diodes (LEDs) and laser diodes (LDs) with emission wavelength in the visible and near ultraviolet spectral regimes attract increasing interest [1]–[5]. Particularly, the ternary InGaN QWs are widely used in blue- and green-emitting LEDs, which have great potential to serve as the next-generation illumination devices [6]–[10]. However, it is still challenging to achieve high performance InGaN QWs, especially in the green and longer wavelength region, due to: 1) high threading dislocation densities in InGaN QWs, originating from the lacking of lattice-matched substrate; 2) phase separation in high-In content InGaN QWs; and 3) severe charge separation due to the existence of internal electrostatic field in c-plane InGaN QWs. InGaN QWs grown along c-plane sapphire substrate contain both spontaneous and piezoelectric polarizations, which induces significant reduction of the electron-hole wavefunction overlap \(\Gamma_{e,h}^{c}\) in InGaN QWs [11]. In order to extend the emission wavelength of InGaN QWs toward green and longer wavelength, InGaN QWs with higher In-content and wider QW thickness are needed, both of which lead to larger electrostatic field in InGaN QW and more severe charge separation. This results in significant reduction of electron-hole wavefunction overlap \(\Gamma_{e,h}^{c}\) and reduction of the internal quantum efficiency [12].

Recently, several approaches have been proposed to suppress the charge separation issue and enhance the electron-hole wavefunction overlap \(\Gamma_{e,h}^{c}\) in c-plane InGaN QWs by using the large overlap QW design such as: 1) staggered InGaN QW [8], [12]–[16], 2) type-II InGaN-GaNAs QW [17]–[19], 3) InGaN QW with \(\delta\)-AlGaN layer [20], [21], 4) strain-compensated InGaN-AlGaN QW [22], 5) InGaN-delta-InN QW [23], [24]. The purpose of using these novel quantum well designs is to enhance the electron-hole wavefunction overlap \(\Gamma_{e,h}^{c}\) and thus to improve the spontaneous emission radiative recombination rate \(R_{sp}\) via engineering the energy band lineups of InGaN QWs.

In this study, we performed the comprehensive studies of the interdiffused InGaN QWs [25]–[27]. The optical properties of the interdiffused InGaN QWs emitting in both blue and green spectral regimes with various interdiffusion lengths \(L_d\) were compared to that of the conventional InGaN QWs. Note that the approach based on the novel QW designs requires modifications and optimizations of the epitaxial recipe of the InGaN QWs in order to grow different types of QWs [12]–[23]. In contrast, interdiffusion based on rapid thermal annealing (RTA) approach is a post-growth procedure for engineering the QW shapes widely implemented in InGaAs/GaAs heterostructures [28]–[30], which avoids the need for modification in the epitaxial recipe for lasers and LEDs.

II. IN/GA INTERDIFFUSION IN INGAN QWS

Interdiffusion based on RTA is a cost-effective approach to improve the optical and electrical properties of as-grown InGaN/GaN QWs, especially for the InGaN QWs with high-In content [31]. During the rapid thermal annealing process, the interdiffusion of indium (In) and gallium (Ga) atoms at the interface of InGaN QW and GaN barrier takes place, where indium atoms in InGaN QW diffuse into GaN barrier region and gallium atoms in GaN layer diffuse into InGaN QW region. The interdiffusion of indium and gallium between InGaN QWs and GaN barriers significantly modifies the energy band lineups for InGaN QWs, which leads to the shift of both electron and hole wavefunctions, resulting in enhancement of the electron-hole wavefunction overlap \(\Gamma_{e,h}^{c}\). Note that the...
interdiffusion length ($L_{d}$) has a significant effect on the energy band lineups, which could be controlled via the rapid thermal annealing condition [27].

Fig. 1 plots the schematics of (a) the conventional InGaN QW and (b) the interdiffused InGaN QW structures, both of which are surrounded by GaN barrier layers. From Fig. 1(a), the conventional InGaN QW contains uniform indium content across the whole InGaN QW region, resulting in sharp energy band lineups at the InGaN/GaN interfaces. From Fig. 1(b), the interdiffused InGaN QW contains an error-function like indium content profile at the InGaN/GaN interfaces [25], resulting in smooth energy band lineups at the InGaN/GaN interfaces. The thickness of the transition layer (as shown in Fig. 1(b)) is defined as two times of the interdiffusion length ($2L_{d}$). Note that the interdiffusion length could be engineered by controlling the RTA conditions such as the RTA temperature, the RTA duration, the interdiffusion length could be engineered by controlling the RTA ambient [27].

The smooth energy band lineups for the interdiffused InGaN QWs push the electron and hole wavefunctions toward the center of the QW region, resulting in enhanced electron-hole wavefunction overlap ($\Gamma_{e-h,\parallel}$). According to Fermi’s Golden Rule, the rate of interband transition is proportional to the square of electron-hole wavefunction overlap ($\Gamma_{e-h,\parallel}^2$) [32]. Thus, the interdiffused InGaN QWs are expected to show higher spontaneous emission radiative recombination rate ($R_{sp}$) than that of conventional InGaN QWs.

III. SIMULATION METHOD

The calculation of the energy band structure of the InGaN QWs is based on a self-consistent 6-band $k \cdot p$ method for wurtzite semiconductors [22], [32], [33]. In the calculation, the model takes into account the valence band mixing, strain effect, spontaneous and piezoelectric polarizations, and carrier screening effect [22]. The coupling between the conduction band and valence band is negligible and not considered here, as the band gap in InGaN QW is relatively large. The calculation of the spontaneous emission radiative recombination rate ($R_{sp}$) in InGaN QWs includes both transverse electric (TE) and transverse magnetic (TM) polarization components, and it is obtained by averaging the momentum matrix elements of TE polarization ($|M_{TE}|^2$) and TM polarization ($|M_{TM}|^2$) as $|M_{sp}|^2 = (2|M_{TE}|^2 + |M_{TM}|^2)/3$. The details of the simulation model including band parameters and material properties are based on the data used in [22], [32], and [33]. The parameters involved in the calculation of optical properties for InGaN QWs were obtained from other [34]. For the ternary In$_x$Ga$_{1-x}$N alloy, the corresponding parameters can be calculated by the linear interpolation of parameters from GaN and InN, except for the energy band gap $E_g$, which can be expressed as:

$E_g(\text{In}_x\text{Ga}_{1-x}\text{N}) = x \cdot E_g(\text{InN}) + (1 - x) \cdot E_g(\text{GaN}) - b \cdot x \cdot (1 - x)$

Note that the bowing parameter $b$ is set as 1.4 eV [34], the band offset ratio ($\Delta E_c : \Delta E_v$) of InGaN/GaN is set as 0.7:0.3 in our calculation.

For the interdiffused InGaN QWs, the indium content profile at the GaN-InGaN-GaN interfaces is determined by Fick’s Law [35], as follow:

$$C(z) = \frac{1}{2} C_0 \left[ \text{erf} \left( \frac{h - z}{L_{d}} \right) + \text{erf} \left( \frac{h + z}{L_{d}} \right) \right]$$

where $C_0$ is the initial indium content, $h$ is the QW width, $z$ is the coordinate along the crystal growth direction and $L_d$ is the diffusion length. Note that the diffusion length $L_d$ is determined by both temperature and material properties [36]. The diffusion lengths and coefficients for In/Ga atoms across InGaN interface had been reported in [36]. The diffusion coefficient for In/Ga atoms at InGaN interface [36] was found to follow Arrhenius expression as

$$D = 1.93 \times 10^{11} \exp(-40473/T) \text{Å}^2/\text{sec},$$

where $T$ represents temperature in K. The diffusion length $L_d$ is related to the diffusion coefficient $D$ by the following relation $L_d = 2\sqrt{D \cdot t}$, where $t$ represents the time of diffusion.

The spontaneous emission spectra can be obtained by taking into account all the transitions between the $n$th conduction subband and the $m$th valence subband as follow [22], [29], [33]:

$$r_{nm}^{\text{sp}}(\hbar \omega) = \frac{2g^2\pi}{n_{\text{e}},n_{\text{h}},\text{L}_\text{w}} \sum_{\nu} \sum_{n,m} \int \frac{k_{d}dk_{\nu}}{2\pi} |M_{\nu m}(k_{\nu})|^2 \times \frac{f_{\nu}^\text{<}(k_{\nu}) (1 - f_{\nu m}(k_{\nu})) (\gamma/\pi)}{\left( E_{\nu m}(k_{\nu}) - \hbar \omega \right)^2 + \gamma^2}$$

where $f_{\nu}^\text{<}(k_{\nu})$ and $f_{\nu m}(k_{\nu})$ are the Fermi-Dirac distribution functions for the electrons in conduction band and holes in valence band, $k_{\nu}$ is the in-plane wave vector, $L_w$ is the thickness of the QW, $(M_{\nu m})^2$ is the momentum matrix element between the $n$th conduction subband and the $m$th valence subband. Because of the lacking of symmetry of the energy band lineups for the conduction band and valence band, the transitions between states with unequal quantum numbers $(m \neq n)$ are not zero. In the simulation, all possible transitions between the confined states of the $n$th conduction subbands and $m$th valence subbands are taken into account.

To investigate the improvement of the optical properties for the interdiffused InGaN QWs, the spontaneous emission radiative recombination rate ($R_{sp}$) is calculated by integrating the (2) over the entire frequency range as follow:

$$R_{sp} = \int_0^{\infty} r_{nm}^{\text{sp}}(\hbar \omega) d(\hbar \omega).$$

IV. SIMULATION RESULTS

In this study, we performed the optimization studies of the interdiffused InGaN/GaN QWs emitting in blue and green spec-
It is observed that the overlap from 17.3% to 22.1%.

The QWs with interdiffusion length of for which is used from 1 to increase, the lower con, corresponding to the decrease N (3.5-nm In to 465 nm for N QWs increases from 17.3% N QWs with various In-con- can be achieved [27].

The N QWs with (with ) as a function of peak emission for in- terdiffused InGaN QWs, the effect of emitting at the similar emission wavelength, Fig. 4 plots the electron-hole wavefunction overlap for the interdiffused 3-nm In(Ga) QWs with that of the conventional InGaN QW, the peak emission wavelength blue-shifts from 495 nm for conventional InGaN QW (Ld = 0 Å) to 465 nm for interdiffused InGaN QW (Ld = 10 Å).

In order to compare the optical performance of the interdiffused InGaN QWs with that of the conventional InGaN QWs emitting at the similar emission wavelength, Fig. 4 plots the electron-hole wavefunction overlap \((\Gamma_{\text{e}, \text{h}})\) for the interdiffused 3-nm In\(_{0.25}\) Ga\(_{0.75}\) N QWs with \(x = 0.25\) to \(x = 0.215\) as a function of peak emission wavelength \(\lambda_{\text{peak}}\). It is observed that the overlap \(\Gamma_{\text{e}, \text{h}}\) for the conventional 3-nm In\(_{0.25}\) Ga\(_{0.75}\) N QWs increases from 17.3% to 21% as the peak emission wavelength \(\lambda_{\text{peak}}\) decreases from 495 nm \(x = 0.25\) to 465 nm \(x = 0.215\), which is due to the reduction of the electrostatic field in the InGaN QW with decrease of the In-content. While for the interdiffused 3-nm In\(_{0.25}\) Ga\(_{0.75}\) N QWs, the overlap \(\Gamma_{\text{e}, \text{h}}\) is enhanced from 17.3% to 24.5% with the increase of interdiffusion length \(L_d\) from \(L_d = 0\) Å to \(L_d = 10\) Å, corresponding to the decrease of the peak emission wavelength \(\lambda_{\text{peak}}\) from 495 nm to 465 nm. From Fig. 4, the interdiffused 3-nm In\(_{0.25}\) Ga\(_{0.75}\) N QWs show enhanced electron-hole wavefunction overlap \((\Gamma_{\text{e}, \text{h}})\) as compared to that of the conventional 3-nm In\(_{0.25}\) Ga\(_{0.75}\) N QWs emitting at the same peak emission wavelength.

The spontaneous emission radiative recombination rate \(R_{\text{sp}}\) for the interdiffused 3-nm In\(_{0.25}\) Ga\(_{0.75}\) N QWs with various interdiffusion length \(L_d\) are calculated as compared to that of the conventional 3-nm In\(_{0.25}\) Ga\(_{0.75}\) N QWs with different In-contents \(x\). Note that the conventional 3-nm In\(_{0.25}\) Ga\(_{0.75}\) N QWs are designed such that the peak emission wavelengths are similar to that of the interdiffused 3-nm In\(_{0.25}\) Ga\(_{0.75}\) N QWs. The calculated spontaneous emission spectra for both structures are
shown in Fig. 5 with carrier density $n = 1 \times 10^{19}$ cm$^{-3}$ and temperature $T = 300$ K.

From Fig. 5, the spontaneous emission spectra of the interdiffused 3-nm In$_{0.25}$Ga$_{0.75}$N QWs are significantly enhanced as compared to that of the conventional 3-nm In$_x$Ga$_{1-x}$N QWs emitting at the similar peak emission wavelength $\lambda_{\text{peak}}$. For the conventional 3-nm In$_x$Ga$_{1-x}$N QWs, the spontaneous emission radiative recombination intensities increase as the peak emission wavelength $\lambda_{\text{peak}}$ blue-shifts with the increase of In-content $x$. While for the interdiffused 3-nm In$_{0.25}$Ga$_{0.75}$N QWs, the spontaneous emission radiative recombination intensities increase as the peak emission wavelength $\lambda_{\text{peak}}$ blue-shifts with the increase of interdiffusion length $L_d$.

Fig. 6 shows the spontaneous emission radiative recombination rate ($R_{\text{sp}}$) for both the interdiffused 3-nm In$_{0.25}$Ga$_{0.75}$N QWs and the conventional 3-nm In$_x$Ga$_{1-x}$N QWs as a function of the peak emission wavelength ($\lambda_{\text{peak}}$). The spontaneous emission radiative recombination rate ($R_{\text{sp}}$) for both QW structures increase with the blue-shift of the peak emission wavelength ($\lambda_{\text{peak}}$) in the blue spectral regime. The interdiffused 3-nm In$_{0.25}$Ga$_{0.75}$N QWs shows higher $R_{\text{sp}}$ than that of the conventional 3-nm In$_x$Ga$_{1-x}$N QWs at the similar peak emission wavelength ($\lambda_{\text{peak}}$). For example, at the peak emission wavelength $\lambda_{\text{peak}} = 470$ nm, the $R_{\text{sp}}$ of the interdiffused 3-nm In$_{0.25}$Ga$_{0.75}$N QWs (with $L_d = 8$ Å) is $6.3 \times 10^{25}$ s$^{-1}$ cm$^{-3}$, while the $R_{\text{sp}}$ of conventional 3-nm In$_2$Ga$_{1-x}$N QWs (with $x = 0.22$) is $4.5 \times 10^{25}$ s$^{-1}$ cm$^{-3}$, indicating an improvement of $\sim 1.4$ times.

Fig. 7 shows the comparison of the $R_{\text{sp}}$ for the conventional 3-nm In$_{0.25}$Ga$_{0.75}$N QWs and the interdiffused 3-nm In$_{0.25}$Ga$_{0.75}$N QWs with $L_d = 8$ Å as a function of the carrier density ($n = 0-1 \times 10^{19}$ cm$^{-3}$). Both QWs emit at the peak emission wavelength $\lambda_{\text{peak}}$ of 470 nm. From Fig. 7, the $R_{\text{sp}}$ of both 3-nm In$_{0.25}$Ga$_{0.75}$N QWs and 3-nm In$_{0.25}$Ga$_{0.75}$N QWs with $L_d = 8$ Å increase with the increase of the carrier density. The interdiffused InGaN QW shows enhanced $R_{\text{sp}}$ at different carrier density as compared to that of the conventional InGaN QW. The enhancement of the $R_{\text{sp}}$ for the interdiffused InGaN QWs is due to the engineered energy band lineups with smooth transition layer at the InGaN/GaN interfaces, which leads to the shift of the electron and hole wavefunctions with enhanced electron-hole wavefunction overlap.

The simulation results from this study show the similar trend as the experimental results [27], [36]. As the diffusion length $L_d$ increases, the interband transition wavelength blue-shifts accompanied by the increase in the spontaneous emission radiative recombination rate.

B. Interdiffused Green InGaN QWs

As compared to the blue-emitting InGaN QWs, it is more challenging to achieve high performance green InGaN QWs due to the requirement of thick QW and relatively high In-content for the InGaN QWs [12]. The interdiffused InGaN QWs emitting in the green spectral region are analyzed as improved
QWs with enhanced electron-hole wavefunction overlap. Specifically, the following two structures are calculated and compared: 1) the conventional 3.5-nm In₇ₓGa₉₋ₓN QWs with various In-content (x); and 2) the interdiffused 3.5-nm In₀.₃Ga₀.₇N QWs with various interdiffusion lengths (l_d).

Fig. 8 plots the energy band lineups and electron and hole wavefunctions for the conventional 3.5-nm In₀.₃Ga₀.₇N QW (black dash line) and the interdiffused 3.5-nm In₀.₃Ga₀.₇N QW with interdiffusion length of l_d = 5 Å (red solid line). Similar to the blue-emitting InGaN QWs, the step-function like energy band lineups for the conventional 3.5-nm In₀.₃Ga₀.₇N are transformed to error-function like band lineups for the interdiffused InGaN QW, which pushes the electron and hole wavefunctions toward the center of the QW, resulting in enhanced electron-hole wavefunction overlap $\langle \psi_{el} \rangle$.

Fig. 9 shows the electron-hole wavefunction overlap $\langle \psi_{el, hh} \rangle$ for the conventional 3.5-nm In₀.₃Ga₀.₇N QWs and the interdiffused 3.5-nm In₀.₃Ga₀.₇N QWs as a function of peak emission wavelength $\lambda_{peak}$. We observe a significant enhancement of the overlap $\langle \psi_{el, hh} \rangle$ for the interdiffused InGaN QW compared to that of the conventional one as the blue-shift of the peak emission wavelength $\lambda_{peak}$. At $\lambda_{peak} = 540$ nm, the $\langle \psi_{el, hh} \rangle$ is enhanced by 1.33 times for the interdiffused InGaN QW as compared that of the conventional one. Fig. 10 plots the confined energy states (EC1 and EC2) and the valance bands (HH1, LH1, HH2 and LH2) for the interdiffused 3.5-nm In₀.₃Ga₀.₇N QWs as a function of interdiffusion length $l_d$.

Fig. 11 shows the comparison of the spontaneous emission spectra between the interdiffused 3.5-nm In₀.₃Ga₀.₇N QWs and the conventional 3.5-nm In₀.₃Ga₀.₇N QWs emitting in green spectral region. As the blue-shift of the peak emission wavelength, both interdiffused and conventional QWs show increase in spontaneous emission spectra. Yet the interdiffused InGaN QWs show stronger enhancement as compared to that of the conventional one. This indicates that the interdiffusion allows a significant enhancement of the spontaneous emission radiative recombination rate for the InGaN QWs emitting in the green spectral region. To illustrate this more clearly, Fig. 12 plots the spontaneous emission radiative recombination rate $R_{SP}$ of the interdiffused InGaN QW and the conventional one as a function of the peak emission wavelength. The $R_{SP}$ of the interdiffused InGaN QW increases more rapidly as compared to that of
Fig. 12. Spontaneous emission radiative recombination rate \( R_{sp} \) for the conventional 3.5-nm In\(_{0.3}\)Ga\(_{0.7}\)N QWs and the interdiffused 3.5-nm In\(_{0.3}\)Ga\(_{0.7}\)N QWs as a function of peak emission wavelength (\( \lambda_{p+x+k} \)).

Fig. 13. The spontaneous emission radiative recombination rate \( R_{sp} \) as a function of charge density in green spectral region.

Fig. 14. The \( R_{sp} \) ratio of interdiffused InGaN QWs over the conventional InGaN QWs in (a) the blue spectral regime and (b) green spectral regime.

C. Blue and Green Interdiffused InGaN QWs

In order to compare the optical performances for the interdiffused InGaN QWs emitting at blue and green spectral regimes, Fig. 14 plots the \( R_{sp} \) ratio of the interdiffused and conventional InGaN QWs for both blue [Fig. 14(a)] and green [Fig. 14(b)] wavelength regimes.

In the blue emission, the interdiffused InGaN QW shows enhancement of the \( R_{sp} \) as the wavelength shortens. The enhancement ratio increases from 1.1 times at 494 nm to 1.5 times at 475 nm. Note that the ratio reaches its peak at around 475 nm, which indicates there is an optimized interdiffusion length for the blue-emitting InGaN QWs. In the green emission, the interdiffused InGaN QW shows enhancement of the \( R_{sp} \). The enhancement ratio increases from 1.2 times at 574 nm to 1.8 times at 533 nm. This indicates that the interdiffusion is an effective approach to enhance the optical performance for both blue and green emitting InGaN QWs. The enhancement is more significant for the green-emitting InGaN QWs with high In-content and thick QW. Thus, interdiffusion has great potential to serve as a cost-effective approach to enhance the performance for InGaN QWs LEDs, especially emitting at green and longer wavelength regimes.

V. SUMMARY

In summary, the interdiffused InGaN QWs emitting in both blue and green spectral regimes are comprehensively analyzed. With the error-function like interface band lineups, the interdiffused InGaN QWs show enhanced electron-hole wavefunction overlap as compare to that of the conventional InGaN QWs emitting at similar wavelength. The interdiffusion, which is achievable by rapid thermal annealing, provides a cost-effective approach to engineer the QW band structures for enhancing the QW performance, which has great potential to achieve InGaN QW based LEDs with improved radiative recombination rate and radiative efficiency emitting applicable for solid state lighting.

REFERENCES


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