Photon recycling white light emitting diode based on InGaN multiple quantum well heterostructure

V.V. Nikolaev(a), M.E. Portnoi(a), and I. Eliashevich(b)
(a) School of Physics, University of Exeter, Stocker Road, Exeter, EX4 4QL, UK
(b) GELcore LLC, 394 Elisabeth Ave., Somerset, NJ 08873, USA

A numerical method based on the transfer matrix technique is developed to calculate the luminescence spectra of complex layered structures with photon recycling. Using this method we show a strong dependence of the emission spectra on the optical eigenmode structure of the device. The enhancement of the photon recycling and the LED external efficiency can be achieved by placing the active regions inside single or coupled microcavities.

I. INTRODUCTION

Recently, great interest has been shown in the use of light emitting diodes (LEDs) as a light source for illumination [1]. LEDs offer many potential advantages compared to conventional light sources due to their relatively low energy consumption, long lifetime and high shock resistance.

Currently white-light LEDs use photo-excitation of phosphors to convert the blue light from an InGaN/GaN LED into white light. However, phosphor has a broad emission spectrum, thus, white LEDs based on this principle do not have the maximum possible luminous efficacy.

The aim of this work is to investigate the feasibility and limitations of creating a white LED by integration within the same structure of several semiconductor layers emitting three basic colours. The recent progress in the growth of InGaN-based double heterostructures and quantum wells makes this alloy an ideal material for the LED active regions, due to the wide variety of the energy gaps in the InGaN system, covering frequencies from red to ultraviolet. The working regime of such a device can be achieved by electrical pumping of active layers with the widest bandgap (blue regions), and by making use of re-emission of the light, absorbed by all the active regions (so called photon recycling).

II. MODELLING OF PHOTON RECYCLING

We start by writing an expression for the intensity of spontaneous emission from a quantum well (QW) into a bulk dielectric material. The number of photons with energy within the interval \([\hbar \omega, \hbar \omega + d(\hbar \omega)]\), which are emitted from the surface \(dS\) into the solid angle \(d\Omega_0\) during the time interval \(dt\) is given by

\[
dN = W_0 \frac{\varepsilon \omega^3 c \hbar^2 p_{cv}^2 g_{2D}}{2\pi^2 e^4 m_0^2} \left[ \int_0^{+\infty} \frac{f_e f_h \Gamma}{(E_{2D} + \varepsilon)^2((E_{2D} + \varepsilon - \hbar \omega)^2 + \Gamma^2)} d\varepsilon \right] dt d(\hbar \omega) dS d\Omega_0. \tag{1}
\]

Here \(E_{2D}\) is the energy gap between the electron and hole quantized levels in the QW, \(g_{2D}\) is the reduced two-dimensional density of states, \(f_e\) and \(f_h\) are the electron and hole occupation probabilities, \(\varepsilon\) is the dielectric constant of the material containing the well, \(m_0\) is the free electron mass, and \(\Gamma\) accounts for the interband relaxation and other broadening mechanisms. The squared momentum matrix element \(p_{cv}^2\) is given in the effective mass approximation for deep QWs by [2]

\[
p_{cv}^2 = \frac{p_0^2}{2} (1 + \gamma)
\]

for TE modes, and by

\[
p_{cv}^2 = \frac{p_0^2}{2} [(1 + \gamma) \cos \theta + (1 - \gamma) \sin \theta]
\]

for TM modes. Here \(\theta\) is the angle between the plane-wave propagation direction and \(Z\)-axis (normal to the QW plane), and \(\gamma = (E_{2D} - E_g)/(\hbar \omega - E_g)\), where \(E_g\) is the bandgap energy of the QW material, and \(p_0\) is the interband momentum matrix element.
We will use the transfer matrix technique in the plane waves basis to analyse the optical properties of layered structures. The plain-wave mode interaction with the QW is described by a QW transfer matrix. If the quantum well width is much smaller than the light wavelength, the transfer matrix has the form:

$$\tilde{M} = \begin{pmatrix} 1 + Y & Y \\ -Y & 1 - Y \end{pmatrix}. $$

Here $Y$ is defined by the two-dimensional QW optical susceptibility $\chi_{2D}$:

$$Y = \frac{2\pi k_0^2}{k_z} \chi_{2D}, \quad (2)$$

where $k$ is the wavevector of light, $k_0 = \omega/c$, and $\chi_{2D}$ for a single-subband QW is given by:

$$\chi_{2D} = \frac{\hbar^2 e^2 \rho_{\text{rel}}^2 g_{2D}}{m_0^2} \int_0^{+\infty} \frac{1 - f_h - f_e}{(E_{2D} + \varepsilon)^2(E_{2D} + \varepsilon - \hbar\omega - i\Gamma)} d\varepsilon.$$

To calculate the rate of photon extraction from a complex structure we consider the interference of all possible processes, resulting in light emission out of the structure. These calculations require a knowledge of the amplitude transmission and reflection coefficients $t_i$ and $r_i$ for the structure part on the left of the QW, and similar coefficients $t_r$ and $r_r$ for the structure part on the right of the QW. We also need to know the transmission and reflection coefficients $t_{QW}$ and $r_{QW}$ for the quantum well itself. Each of these coefficients can be obtained from the corresponding transfer matrix.

Let us derive, for example, the power, emitted from the right side of the structure. A photon emitted inside the structure can be transmitted directly to the outside medium, or it can be consecutively reflected from the right and left sides of the structure and finally will be transmitted outside and so on. The outgoing electric field, resulting from all these processes is given by the sum:

$$E_{r \rightarrow r} = t_r + r_r t_r + r_r r_r^* r_r + \ldots = \frac{t_r}{1 - r_r^* t_r},$$

where the star in $r_r^*$ indicates, that this coefficient includes the reflection from the emitting QW.

Similarly, photons emitted inside the structure to the left can undergo multiple reflections and eventually escape from the structure to the right. These processes give a second part of the external field:

$$E_{l \rightarrow r} = \frac{r_l t_{QW} t_r}{(1 - r_r^* r_r)(1 - r_{QW} r_l)}.$$

Thus, we obtain the expression for the emission from the right side of the structure:

$$dN_{Er} = W_0 \cdot \left| \frac{t_r}{1 - r_r^* t_r} \left[ 1 + \frac{r_l t_{QW}}{1 - r_{QW} r_l} \right] \right|^2 \sqrt{\epsilon_r k_{xr}} \sqrt{\epsilon_0 k_{2D}} dt \cdot d(h\omega) \cdot dS \cdot d\Omega_e, \quad (3)$$

where the ratio $\sqrt{\epsilon_r k_{xr}}/\sqrt{\epsilon_0 k_{2D}}$ accounts for the change in solid angle that is due to refraction for plane waves. The indices $r$ and $0$ are related to the outside medium and the layer containing the QW, respectively. To obtain the total density of the external light intensity we have to sum over all QWs and integrate Eq.(3) over the external solid angle $\Omega_e$.

If we neglect the reflection from the QW by substituting $t_{QW} = 1$ and $r_{QW} = 0$ into Eq.(3), the formula for extraction becomes analogous to one obtained using the source-term method.

For the quantitative description of the recycling process, we have to calculate the rate of absorption by the quantum well $QW_a$ of the spontaneous emission from the other well, $QW_e$. Thus, we need to know the induced electric field at the position of $QW_a$. The power flux balance shows, that the rate of absorption of emitted photons by the unit area of surface of $QW_a$ is given by:

$$W_a = -\frac{1}{2} \sqrt{\epsilon_z} |E|^2 \text{Re}(Y_{QW_a}) W_{QW_e}, \quad (4)$$

where $E$ is the complex amplitude of the field at the $QW_a$ location, $Y_{QW_a}$ is defined by Eq.(2), and $W_{QW_e}$ is defined by Eq.(4). The induced field can be calculated in a similar fashion as it was done for extraction:

$$E = \frac{t(1 + r_{a\alpha}^*)}{(1 - r^* r_{a\alpha})(1 - r_{a\alpha}^* r_{a\alpha})} \left( 1 + \frac{t_{QW_e} r_{er}}{1 - r_{er} r_{QW_e}} \right).$$
where \( r_{al} \) (\( r_{er} \)) is a reflection coefficient for the part of structure to the left (right) of the well \( \text{QW}_a \), \( r_{er} \) is a reflection coefficient for all the layers to the right of the well \( \text{QW}_c \), and the coefficients \( r \) and \( t \) correspond to the part of the structure between \( \text{QW}_c \) and \( \text{QW}_a \). Here we assume that \( \text{QW}_a \) is on the left of \( \text{QW}_c \). The formula for the opposite case is similar.

One of the channels for the photon to escape from the recycling process is to be absorbed in a metallic mirror. Placing such a mirror onto the left side of the structure, and denoting the reflection coefficient of the left part of the structure, excluding the mirror, as \( r_l \), the reflection coefficient from the mirror as \( r_m \), the reflection coefficient for the wave incident from the mirror as \( r_s \), and the transmission coefficient from the \( \text{QW} \) to the mirror as \( t \), we obtain the following expression for the number of absorbed photons:

\[
dN_m = W_0 \left( 1 - |r_m|^2 \right) \frac{t}{|1 - r_l r_c^* (1 - r_m r_s)|} \left( 1 + \frac{r_r t_{QW}}{1 - r_r t_{QW}} \right)^2 dR \cdot dS \cdot d\Omega_0.
\]  

(5)

We restrict our consideration by relatively low pumping levels, which are typical for the diode operation regime. Thus, the expression for the electron density \( \rho_e \) in the one-subband \( \text{QW} \) can be simplified (temperature is measured in energy units):

\[
n = \frac{m_e}{\hbar^2 \pi} T \ln[1 + \exp(\mu_e/T)] \approx \frac{m_e}{\hbar^2 \pi} T \exp(\mu_e/T),
\]

and the occupation probabilities can be expressed as:

\[
f_e = \frac{1}{1 + \exp(\varepsilon_e/\mu_e)} \approx \exp(\mu_e - \varepsilon_e/\mu_e) \approx n \frac{\hbar^2 \pi}{m_e T} \exp(-\varepsilon_e/\mu_e),
\]

\[
f_h = \frac{1}{1 + \exp(\varepsilon_h/\mu_h)} \approx \exp(\mu_h - \varepsilon_h/\mu_h) \approx p \frac{\hbar^2 \pi}{m_{hh} T} \exp(-\varepsilon_h/\mu_h),
\]

where \( m_e \) (\( m_{hh} \)) and \( \mu_e \) (\( \mu_h \)) are the electron (heavy hole) effective mass and quasi-Fermi-level, respectively. Under the low-pumping assumption we can rewrite \( W_0 \) in the form:

\[
W_0 = \frac{\omega^2 \hbar^2 c^2 p_c^2 q_2 D}{2 \pi^2 c^3 m_0^2} \left[ \int_0^{+\infty} \frac{\Gamma \exp(-\varepsilon/\mu_e) d\varepsilon}{(E^2D + \varepsilon)^2 ((E^2D + \varepsilon - \hbar\omega)^2 + \Gamma^2)} \right] \cdot np.
\]

(6)

The total rate of light emission into the external medium can be obtained by substituting Eq.6 in Eq.5 and integrating Eq.5 over the external solid angle and photon energies. As a result the rate of radiative recombination depends on the carrier densities as \( R_{ext} = E \cdot np \).

Due to the large photon energy separation between different colours, the absorption rate for the short-wavelength light is independent of the carrier density in the narrow-gap \( \text{QW} \). For the light emitted from \( \text{QW}_c \) the rate of absorption in \( \text{QW}_a \) is given by the expression \( R_{abs} = A \cdot n_p \cdot c_p \), where \( A \) is calculated by integrating Eq.5 over the total solid angle of emission and over the photon energies. Here we assume that all \( \text{QWs} \) are embedded in layers of dielectric material with the refractive index equal to the highest one in the real structure. This trick allows as to handle the interaction of active layer with the evanescent wave. However, introducing several sufficiently thin layers with high dielectric constant does not alter the optical properties of the structure.

A similar relation holds for the rate of absorption in a metallic mirror: \( R_m = M \cdot np \). Coefficient \( M \) is obtained by substituting Eq.6 in Eq.5 and integrating the result over the solid angle and photon energies. Note, that due to charge-neutrality the electron and hole densities in each \( \text{QW} \) are equal to each other, \( n = p \).

The steady-state carrier densities are given by the balance between generation of electron-hole pairs in the \( \text{QWs} \) and their recombination, both radiative and non-radiative. The generation processes include electric current pumping of the blue \( \text{QWs} \) and the re-absorption of emitted light throughout the structure. The recombination output goes to external emission and internal losses, which we treat as absorption in the other \( \text{QWs} \) and non-radiative recombination in the given \( \text{QW} \). In our approach this leads to the following equation for each \( \text{QW} \):

\[
R_i = E_i n_i^2 + A_i n_i^2 + M_i n_i^2 + n_i / \tau_i,
\]

where \( R_i \) is the pumping rate and \( \tau_i \) is the non-radiative recombination time. Assuming that \( \tau_i \) does not depend on the carrier density, we get:
\[ n_i = \sqrt{\frac{(1/\tau_i)^2 + 4(A_i + E_i + M_i)R_i - 1/\tau_i}{2(A_i + E_i + M_i)}}. \]  

Then the external efficiency for each QW can be obtained as

\[ \eta_i = \frac{E_i n_i^2}{(A_i + E_i + M_i)n_i^2 + n_i/\tau_i}. \]

## III. RESULTS AND DISCUSSION

We used the numerical method described above to investigate a number of different types of structures. It was revealed, that the emission spectra and external efficiencies depend drastically on the active layer positions and on the mode structure of the device. Figure 1 shows the calculated spontaneous emission for a structure, which represents GaN microcavity containing three QWs and covered on one side by a metallic mirror. The width of the GaN layer and the energies of interband transitions in red, green and blue QWs are chosen in such a way that the structure can be regarded as the $5\lambda/2$ resonator for the normally propagating red-light waves, $6\lambda/2$ resonator for the green-light waves and $7\lambda/2$ resonator for the blue-light waves. We placed each QW in an antinode of a resonant mode, associated with the QW colour. Our calculations were performed for different times of non-radiative recombination ranging from 1 ns to 10 ns, and we assumed the carrier density in blue QW to remain constant. One can see, that if the internal losses are high, the recycling efficiency is low and a blue light only is emitted. When the internal efficiency increases, the emission from the optically pumped QWs becomes comparable with the blue-light intensity. The green-light intensity usually remains smaller than both blue and red because of the strong re-absorption in the red active region. However, this is beneficial for the white-light generation, because of the high sensitivity of the human eye in the green region of the spectrum. If high non-radiative loses are present, the recycling process can be enhanced by introducing more red and green QWs in the structure and by building a Bragg reflector for the blue wavelength. A possible way to enhance the external efficiencies of all three colours is to place the active regions into coupled microcavities.

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FIG. 1. Spontaneous emission spectra from a microcavity, containing three quantum wells. The non-radiative recombination times are 1 ns (solid line), 5 ns (dashed line) and 10 ns (dot-dashed line). The carrier density in the blue QW is the same for all three curves.