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Theoretical Analysis of Photo-Recycling Effect on External Quantum Efficiency Considering Spatial Carrier Dynamics

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We discuss the effect of photo-recycling effect on the external quantum efficiency of radiation (EQE) in semiconductors, considering spatial carrier dynamics. We calculate carrier distribution and EQE for both w/ and w/o photo-recycling term, and clarify the effect of photo-recycling. We find that the photo-recycling keep high concentration of photo-excited carrier in the sample and enhances the EQE beyond the light extraction efficiency. However, in strong photo-recycling condition, the EQE enhancement is suppressed by the effective increase in the carrier diffusion length. Our analysis for the photo-recycling effect paves the way for accurate evaluation of crystal quality of semiconductors from the EQE.

1. Introduction

Photoluminescence (PL) measurement is a powerful tool for the evaluation of crystal quality of semiconductor samples. Recently, we have firstly succeeded in quantifying the absolute value of the EQE of a GaN wafer by utilizing the omnidirectional photoluminescence (ODPL) spectroscopy, and estimated the IQE from the absolute EQE [1-2]. The values of EQE and IQE have direct relation with the concentration of the non-radiative recombination centers (NRCs) such as $V_{Ga}V_N$ divacancies in GaN-based materials [3-4]. Therefore the measurement of the quantum efficiencies utilizing ODPL spectroscopy has attracted much attention as a promising method to quantify the NRC concentration. However, in the interpretation of the EQE, the effect of "photo-recycling" is still a matter of debate.

The EQE is given by the ratio of the PL intensity to the absorbed pumping light intensity. When the radiative recombination becomes dominant because of the low NRC concentration, the photo-recycling phenomenon, that is, the self-absorption and the re-emission of photon, plays a significant role in the PL intensity [1,5]. This photo-recycling strongly depends on the carrier distribution in the sample. However, the relation between the photo-recycling effect and the spatial carrier dynamics has not been investigated well. In this work, we numerically calculate the carrier distribution and the PL intensity in semiconductor samples using the drift-diffusion based transport model, and discuss the photo-recycling effect on the EQE.

2. Model and Method

For simplifying the equation for carrier dynamics, we consider a bulk sample whose area of photo irradiation is enough large so that the carrier distribution is almost uniform along the plane parallel to the sample surface (y-z plane). In addition, we assume an N-type sample whose dopant concentration N_0 is larger than the NRC concentration N_t. In a weak photo pumping condition, the continuity equation for hole carrier along the thickness direction (x axis) is reduced to [6],

$$D_h \frac{\partial^2 p(x)}{\partial x^2} = -\alpha \mathcal{I}_{pump} e^{-\alpha x} + N_t C_h p(x) + R_{rad}(x) - R_{self}(x),$$
$$R_{rad}(x) = B n_0 p(x), \quad G_{self}(x) = \frac{\alpha B n_0}{2} \int_0^L \int_0^{\frac{\pi}{2}} p(x') \tan\theta \Big(e^{\frac{-\alpha|x-x'|}{\cos\theta}} + \Gamma(\theta, n_r)\big(e^{\frac{-\alpha|x+x'|}{\cos\theta}} + e^{\frac{-\alpha|2L-x-x'|}{\cos\theta}}\big)\Big) d\theta dx'. \quad (1)$$

Here, $p(x)$ is the hole concentration, D_h is the hole diffusion constant, C_h is the hole capture coefficient by NRC, B is the spontaneous emission coefficient, $n_0 = N_0 - N_t$ is the uniform electron concentration in weak photo pumping condition, α is the absorption coefficient of light, I_{pump} is the intensity of CW pumping light , $\Gamma(\theta, n_r)$ is the Fresnel reflection coefficient depending on the refractive index n_r, L is the sample thickness. G_{self} indicates the generation rate corresponding to the revival of photo-excited carriers by the self-absorption

Parameter	Value
Hole diffusion constant: D_h	0.026 cm^2s^{-1}
Hole caputre coefficient: C_h	7×10^{-7} $cm^{-3}s^{-1}$
N-type Dopant concentration: N_0	1×10^{17} cm^{-3}
NRC concentration: N_t	1×10^{13} cm^{-3}
Pumping light intensity: I_{pump}	1×10^{15} $cm^{-2}s^{-1}$
Absorption coefficient: α	1×10^{5} cm^{-1}
Refractive index: n_r	3.0
Samlple thickness: L	10 μm

Table 1: Caculation parameters for bulk semiconductor sample.

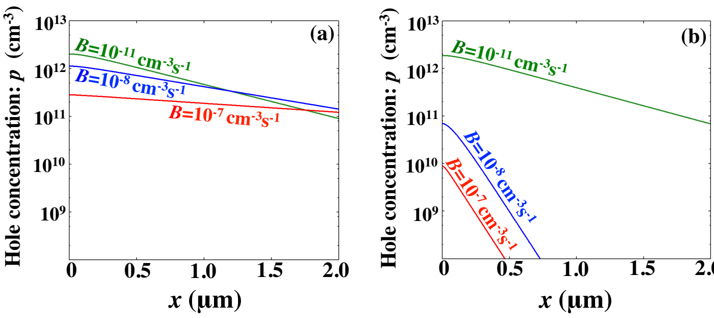

Figure 1: (a) Calculated distribution of hole concentration w/ photo-recycling effect for $B = 10^{-11}, 10^{-8}, 10^{-7}$ $cm^{-3}s^{-1}$. (b) Calculated distribution of hole concentration w/o photo-recycling effect for $B = 10^{-11}, 10^{-8}, 10^{-7}$ $cm^{-3}s^{-1}$.

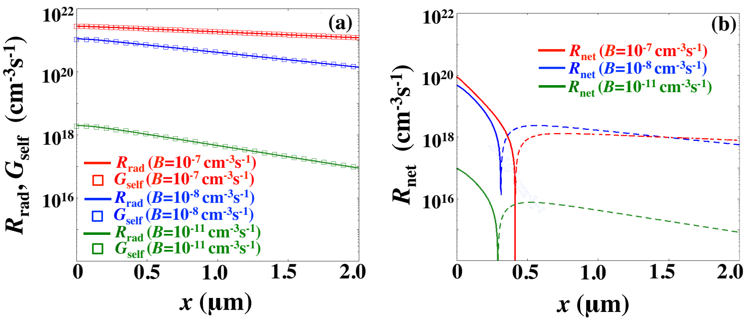

Figure 2: (a) Calculated distributions of $R_{\rm rad}$ and $G_{\rm self}$ w/ photo-recycling effect for B = 10^{-11},10^{-8}, 10^{-7} cm^{-3}s^{-1}. (b) Calculated distribution of net radiative recombination $R_{\rm net} = R_{\rm rad} - G_{\rm self}$ for B = 10^{-11},10^{-8}, 10^{-7} cm^{-3}s^{-1}. The solid and dotted lines indicates positive and negative values, respectively.

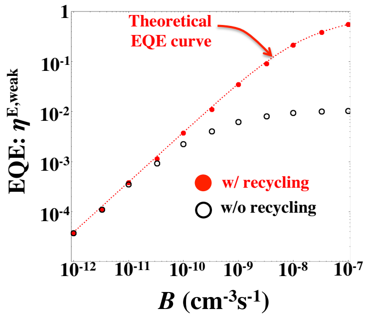

Figure 3: Calculated EQE as a function of B w/- recycling (filled circle) and w/o recycling (open circle). The red dotted line indicates the theoretical EQE curve given by Eq. (2).

process. In this paper, for clarifying the effect of photo-recycling, we calculate hole distributions for the cases w/ and w/o $G_{\rm self}$ term (hereafter we will refer w/ and w/o recycling cases). EQE is calculated as the ratio of the PL intensity $I_{\rm PL}$ to $I_{\rm pump}$. The calculation parameters comparable to GaN crystals [1-4] are listed on Table 1. We consider low NRC concentration condition and change the emission coefficient B from 10^{-12} to 10^{-7} cm^{-3}s^{-1} in order to discuss the photo-recycling effect. The parameter range of B corresponds to the value of IQE from 0.0141 to 0.999.

3. Results

Figures 1 (a) and (b) shows the calculated hole distribution near the photo irradiated surface (x = 0) w/ and w/o recycling for B=10^{-11}, 10^{-8} and 10^{-7} cm^{-3}s^{-1}, respectively. When the radiative recombination is weak (B =10^{-11} cm^{-3}s^{-1}), the distributions of both cases are almost same. However, the difference between them becomes remarkable for large B. The hole concentration rapidly decreases, and the carrier diffusion length λ becomes small with increase in B for w/o recycling case. On the other hand, the decrease of the hole concentration is small, and λ becomes large with increase in B for w/ recycling case. For better understanding of the photo-recycling effect, we show the distribution of $R_{\rm rad}$ and $G_{\rm self}$ for w/ recycling case in Fig. 2(a). As shown in this figure, the values of $R_{\rm rad}$ and $G_{\rm self}$ are close each other. This means that a large part of the photons emitted by the radiative recombination is absorbed in the sample, and it revives photo-excited carriers. Hence, the concentration of photo-exited carriers is kept in w/ recycling case. Figure 2 (b) shows a net radiative recombination rate $R_{\rm net} = R_{\rm rad} - G_{\rm self}$ in w/ recycling case. The solid and dashed lines indicate positive and negative values, respectively. In the surface region, the radiative recombination gets over the self-absorption, and intense emission occurs. Interestingly, $R_{\rm net}$ becomes negative in sample inside region. In the sample inside, $G_{\rm self}$ surpasses $R_{\rm rad}$ because of the absorption of intense luminescence from the surface region. This results in the effective increase of the carrier diffusion length λ [5,6].

Finally, we show the calculated EQE as a function of B for both w/ and w/o recycling cases. The solid line indicates the theoretical curve for the EQE derived in our recent work

$$\eta^{\rm E_{weak}} = \frac{\eta_{\rm self}(\alpha,\lambda)\eta_{\rm opt}(n_{\rm r})Bn_0}{\eta_{\rm self}(\alpha,\lambda)\eta_{\rm opt}(n_{\rm r})Bn_0 + N_{\rm t}C_{\rm h}}. \quad (2)$$

[6],

Here, the surface recombination rate is ignored, and $\eta_{\rm opt}(n_{\rm r})$ is the light extraction efficiency obeying Fresnel's law, $\eta_{\rm self}$ (α,λ) is the light extraction efficiency which gives the fraction of the luminescence reaching to the sample surface overcoming the self-absorption process. The calculated EQE for w/ recycling case agrees well with the theoretical curve. The difference between w/ and w/o recycling cases becomes significant as B increases. As shown in Fig. 1 (b), the concentration of photo-excited carrier is kept by the photo-recycling effect in w/ recycling case, and the PL intensity keep increasing with increase in B. Meanwhile, in w/o recycling case, photo-excited carrier drastically decreases with increase in B because of the strong radiative recombination without photo-recycling, and the EQE saturates to the value of the extraction efficiency. Of particular note that the increase of EQE in w/ recycling case is suppressed for large B (B=10^{-8}-10^{-7} cm^{-3}s^{-1}). The strong photo-recycling effect effectively increases the diffusion constant of the carrier, and the strong diffusion decreases the carrier concentration near the surface (compare the distributions for B=10^{-11} and B=10^{-7} in Fig.1 (a)). As a result, the extraction efficiency originating from the self-absorption $\eta_{\rm self}$ decreases, and the increase of the EQE is suppressed.

4. Conclusion

In this paper, we theoretically investigate the photo-recycling effect on EQE based on drift-diffusion based carrier transport model. The photo-recycling effect keep high concentration of photo-excited carrier and enables intense PL from the sample. Meanwhile, the strong photo-recycling promote carrier diffusion, and it suppresses the EQE increase.

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