A NUMERICAL MODEL FOR COMPUTING THE EMISSION SPECTRUM IN TIME-RESOLVED PHOTOLUMINESCENCE EXPERIMENTS

Steven M. Durbin[1], Dean H. Levi[2] and Jeffery L. Gray[1]
[1]*School of Electrical Engineering, Purdue University, West Lafayette, IN 47907-1285*
[2]*National Renewable Energy Laboratory, 1617 Cole Blvd., Golden, CO 80401*

Abstract

A detailed numerical model is presented for computing the luminescence spectrum in a semiconductor layer which properly accounts for the effects of photon recycling. The resulting model is incorporated into a 1-D drift-diffusion simulation package, and used to simulate time-resolved photoluminescence (TRPL) experiments on AlGaAs/GaAs/AlGaAs double heterostructures in order to extract carrier recombination and transport information. Results of initial simulations are compared with measured spectra to verify the accuracy of the model.

I. INTRODUCTION

A common method for determining minority carrier lifetimes in compound semiconductors is time-resolved photoluminescence (TRPL), which measures the decay of photoluminescence intensity in response to pulsed laser excitation. The effects of re-absorption of photons emitted via radiative recombination (commonly referred to as "photon recycling") have been shown to significantly affect the measured lifetime in direct-gap semiconductors such as GaAs when radiative recombination is dominant or nearly dominant [1]. In order to permit the investigation of the physical processes underlying the experimental technique, a detailed numerical model for the emission intensity has been incorporated into a complete 1-D numerical semiconductor simulation package developed at Purdue University. Simulations of AlGaAs/GaAs/AlGaAs double heterostructures (DHs) are performed in order to explore the sensitivity of the measurement to various device and material parameters.

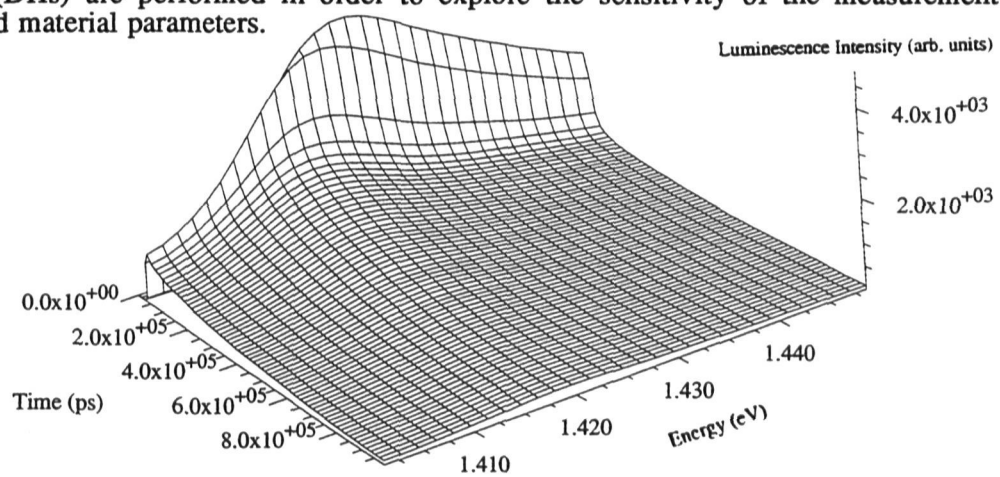

Fig. 1: Experimental time-resolved photoluminescence spectrum of a 10 μm DH.

Because the near-bandedge absorption coefficient increases with photon energy, self-absorption will shift the externally observed emission peak towards lower energies. Thus, as carriers diffuse away from the front surface after the excitation pulse, an increasing fraction of the higher-energy portion of the luminescence will be reabsorbed, resulting in a red shift of the observed spectrum with time. Such an effect has been observed experimentally in AlGaAs/GaAs/AlGaAs double heterostructures (Fig. 1), and simulation results predict similar behavior. Although TRPL is typically used to extract minority carrier lifetimes, the temporal behavior of the peak energy can provide information about the minority carrier diffusivity as well. Thus, an accurate numerical model for TRPL can provide insight into both carrier recombination and transport.

II. NUMERICAL APPROACH

A. Solution of Semiconductor Equations

A one-dimensional finite difference/ finite box discretization technique is applied to the self-consistent solution of the Poisson equation and the electron and hole current continuity equations. Newton-Raphson iteration is employed to solve the resulting set of coupled nonlinear partial differential equations for the electrostatic potential as well as hole and electron concentrations at discrete mesh points within the modeled device. The self-absorption of photons emitted through radiative recombination ("photon recycling") is accounted for by calculation of an effective generation rate after the method of Kuriyama et. al. [2], which is introduced through the generation term in both current continuity equations [3].

B. Numerical Emission Intensity Model

The expression for luminescence intensity begins with assuming rotational symmetry in a cylindrical geometry (r, ϕ, z) having infinite transverse dimensions; the device geometry is taken to vary along the z axis. Assuming infinite transverse dimensions will not affect the accuracy provided that the physical dimensions of the device being modeled are greater than the inverse absorption coefficient corresponding to the emitted photon energy. Photons are assumed to be emitted via radiative recombination throughout the 3-D cylindrical volume, and incident upon a line through the origin (r=0, ϕ, z). A 1-D expression is obtained by assuming that there is no transverse variation in physical variables (i.e. p, n, and V), so that the photon path lengths may be written in terms of a longitudinal component along the z-axis. Considering a differential slab of thickness dz' emitting a flux of $\Phi_0 = 0.5\ B\ [n(z')p(z') - n_i^2]dz'$ towards the front surface (z' = 0), the fraction of photons incident on the interior surface in a differential solid angle $d\Omega$ is given by [4]:

$$d\Phi = \Phi_0 \exp(-\alpha z'/\cos\theta)\, d\Omega/2\pi = \Phi_0 \exp(-\alpha z'/\cos\theta)\sin\theta\, d\theta\, d\phi\, /2\pi \qquad (1)$$

where θ is the angle between the emission ray and the z-axis. The expression includes self-absorption through the exponential decay term, where α is the absorption coefficient. The rotational symmetry of the chosen geometry permits integration over the angular coordinate ϕ, resulting in a factor of 2π. Integrating over the angle θ by making the variable substitution $u = \alpha z'/\cos\theta$, equation 1 becomes:

$$\Phi_{incident} = \Phi_0\, \alpha z' \int_{\alpha z'}^{\infty} \frac{e^{-u}}{u^2} du = \Phi_0\, E_2(\alpha z') \qquad (2)$$

where E_2 is a member of the family of functions known as exponential integrals [5]. The equation differs from the simple exponential expression commonly used to describe absorption ($\Phi = \Phi_0\, e^{-\alpha z'}$) because the majority of photons incident on the interior surface travel further than z' (since the emission is isotropic rather than collimated), resulting in greater absorption of the total flux.

In order to provide the external PL spectrum due to radiative recombination everywhere within the device layer, the contribution from angles greater than the critical angle of reflection (θ_c) must be subtracted from equation 2, and the result multiplied by the polarization-averaged transmissivity of the interface (1 - R). Equation 2 must also be integrated over the entire device width (w), and integrated over energy [6]:

$$\Phi_{emission} = \int_0^{\infty} dE\, \hat{\gamma}(E) \int_0^{w} dz' \frac{B}{2}\left[n(z')\, p(z') - n_i^2\right] \Big\{ (1 - R_f)\left[E_2(\alpha(E)\, z') - \cos\theta_c\, E_2\left(\alpha(E)\, z'/\cos\theta_c\right)\right]$$

$$+ \sum_{k\ even,\ >0} (1 - R_f)\, R_b^{k/2}\, R_f^{k/2} \left[E_2(\alpha(E)\,[kw + z']) - \cos\theta_c\, E_2\left(\alpha(E)\,[kw + z']/\cos\theta_c\right)\right]$$

$$+ \sum_{k\ odd,\ >0} (1 - R_f) R_b^{(k+1)/2}\, R_f^{(k-1)/2} \left[E_2(\alpha(E)\,[(k+1)w - z']) - \cos\theta_c\, E_2\left(\alpha(E)\,[(k+1)w - z']/\cos\theta_c\right)\right] \Big\}$$

where $\hat{\gamma}(E)$ represents the normalized van Roosbroeck- Shockley representation of the spontaneous emission spectrum [2]. In the above equation, k is the number of reflections between interfaces before a photon either escapes or is reabsorbed. To completely account for all spontaneous emission

photons, an infinite number of reflections (k) is required. However, numerically only about 10 reflections are required to obtain a reasonable degree of accuracy, depending on the geometry.

C. Relation to Photon Recycling

In 1-D, the generation rate due to band-to-band absorption of photons is given by:

$$G(z) = -\frac{d\Phi}{dz}.$$

Using this relation on equation 2, an effective photon recycling generation rate can be computed [6]:

$$G_{pr}(z) = -\Phi_0 \frac{d}{dz} E_2(\alpha z) = \alpha \Phi_0 E_1(\alpha z)$$

which is equivalent to the general expression presented by Kuriyama, et. al. [2]. Thus, the total emission (including emission from the front surface, and loss to the substrate) can be expressed in terms of the effective photon recycling generation rate:

$$\Phi_{emission} = \int_{slab} [R_{radiative}(z) - G_{pr}(z)]\, dz$$

In our implementation, self absorption/ photon recycling is incorporated into the solution by computing the effective generation rate and adding it to the external optical generation rate in the current continuity equations. The emission spectrum is then computed as a post-processing step after each iteration so that it is self-consistent with the current solution.

III. SIMULATION RESULTS

In order to determine the quantitative effects of the minority carrier diffusivity on the observed time-dependent PL spectrum, a series of simulations were performed on a 10 μm GaAs DH with a doping of $n \sim 1.3 \times 10^{17}$ cm^{-3}. The absorption coefficients were obtained from transmission measurements on an identical series of structures with various GaAs layer thicknesses [7]. Fig. 2 shows the calculated time-dependent PL spectrum for the structure assuming a bulk nonradiative lifetime of 1.3 μs, a radiative recombination coefficient of 2.45×10^{-10} cm^3/s, and a minority hole

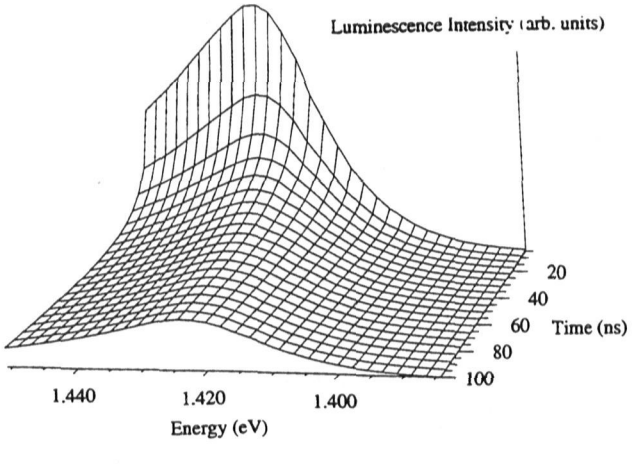

Fig. 2: Calculated time evolution of the PL spectrum of the 10 μm GaAs DH.

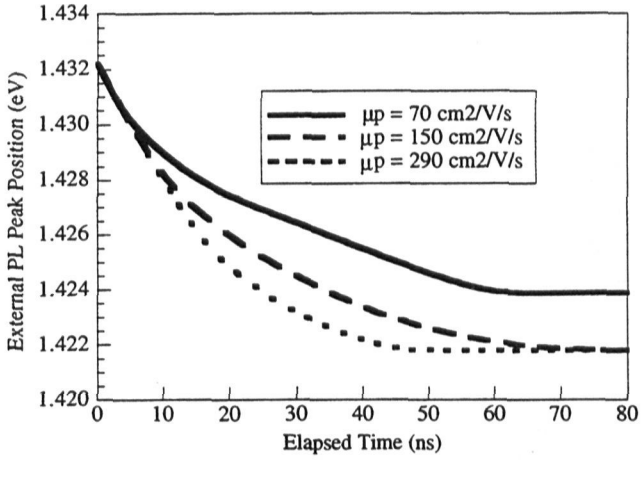

Fig. 3: Comparison of PL spectra for different values of hole mobility.

mobility of 70 cm^2/V/s. As can be seen, the calculated behavior is similar to the experimental results plotted in Fig. 1. The predicted external PL peak as a function of time is plotted in Fig. 3 for minority hole mobilities of 70, 150 and 290 cm^2/V/s. As expected, the peak energy shifts much more quickly with larger values of the mobility, as diffusion moves the excess carriers further away from the front surface before they can recombine.

Initial simulations of the measurement in Fig. 1 did not predict quite as large of a red shift, even with very large values of minority hole mobility. In order to examine this in more detail, steady state PL was performed on the same structure. The results of the best fit are plotted in Fig. 4 (as "predicted PL"), along with the experimental data. As in the TRPL simulations, the predicted external PL peak is approximately 5 meV bluer than the experimental data. However, by assuming a flat excess carrier profile, the E_2 integral expression may be integrated and used to remove the effects of self-absorption on the PL peak. Working backwards in such an approximate fashion results in a reasonable fit to the peak energy, also plotted in Fig. 4 (as "shifted experimental") along with the van Roosbroeck-Shockley (VRS) relation. The disagreement of the "shifted experimental data" curve with the VRS curve away from the peak suggests that either the absorption model or the emission expression may need further refinement. The lack of agreement to the lower energy side of the VRS curve, however, provides a possible explanation for the lack of agreement between the experimental and simulated PL curves, as that energy region overlaps the experimental PL peak.

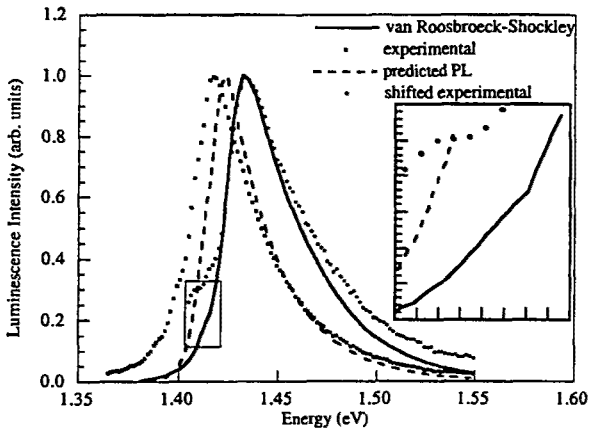

Fig. 4: Experimental steady-state PL of the 10 μm DH. The highlighted region shows where the absorption model accuracy leads to difficulty in predicting external PL.

IV. SUMMARY

A detailed numerical model for computing the emission spectrum in time-resolved photoluminescence experiments has been incorporated into a 1-D semiconductor simulation package. The calculation of the photoluminescence (PL) is performed self-consistently with calculation of the photon recycling generation rate, thereby automatically accounting for the effects of self-absorption on the externally observed emission spectrum. While detailed modeling of the decay of PL intensity provides information with regard to recombination processes, the temporal behavior of the PL spectrum depends on the minority carrier diffusivity as well. Thus, detailed modeling of the TRPL experiments can provide information about both minority carrier transport and recombination.

REFERENCES

[1] R.K. Ahrenkiel, D.J. Dunlavy and T. Hanak, J. of Appl. Phys. **64**, 1916 (1988).
[2] T. Kuriyama, T. Kamiya and H. Yanai, Jpn. J. Appl. Phys. **16**, 465 (1977).
[3] S.M. Durbin and J.L. Gray, IEEE Trans. Elec. Dev. **41**, 239 (1994).
[4] E. Velmre, B. Freiden and A. Udal, Phys. Scr. **24**, 468 (1981).
[5] W. Gautschi and W.F. Cahill, in Handbook of Mathematical Functions, M. Abramowitz and I.A. Stegun, eds., Dover Publications, Inc., New York, 227 (1970).
[6] S.M. Durbin, J.L. Gray, M.P. Patkar and M.S. Lundstrom, Conf. Rec. OE/Lase '94, SPIE **2146**.
[7] G. B. Lush, Ph.D. Thesis, Purdue University (1992).

The computational development work was supported by the NASA Graduate Research Fellowship Program (author S.M.D.). The research at NREL was supported by the Department of Energy under contract number DE-AC02-83CH10093.