

Connecting recombination rates with carrier density via quasi-steady-state and transient photoluminescence in GaAs

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Abstract — The characterization of defects and their impact on recombination rates in solar cells is crucial for evaluating and improving photovoltaic performance. Injection-dependent transient photoluminescence (PL) spectroscopy is a sensitive and commonly used technique for investigating recombination rates in semiconductors. However, determination of the photoexcited carrier density during the lifetime experiment and making comparisons with steady-state measurements can be challenging. We employ a new dynamic calibration approach, which relies on a combination of quasi-steady state and transient PL measurements, to obtain the lifetime as a function of charge carrier density in a GaAs/GaInP heterostructure. We find that detection constraints limit the utility of the experiment in its current configuration.

I. INTRODUCTION

Photovoltaics are the fastest growing renewable energy source, driven by improved performance and reduced cost. However, defects in photovoltaic materials continue to limit conversion efficiencies. Even in GaAs, which is the active material in the most efficient single-junction solar cell, defects contribute to losses via nonradiative recombination. Hence, understanding the complex behavior of various defects can help us develop better photovoltaic devices.

When a short pulse of light generates excess electrons Δn and holes Δp in a semiconductor, several physical processes drive the system back toward equilibrium. These recombination mechanisms can be characterized by lifetimes τ_n and τ_p that depend on the photoexcited carrier densities. For example:

$$\frac{d\Delta n}{dt} = \frac{\Delta n(t)}{\tau_n(\Delta n)} \quad (1)$$

Lifetimes are not difficult to measure – we just need the slope of transient photoluminescence (PL) decay curve after the laser is turned off. But since the carrier density changes throughout this process, the lifetime is steadily changing too.

In this project, we seek to evaluate the rate of recombination in a GaAs/GaInP double heterostructure and connect it with the underlying carrier density. We use a calibration procedure developed by Zhu *et al.* [1] and presented at the 50th Photovoltaic Specialists Conference. [2] Density-dependent lifetime measurements enable us to characterize the impact of defects because defect-related recombination dominates at low densities while filling limits the impact of defects at high density. A quantitative understanding of density-dependent defect recombination and filling can facilitate better device design and operational modeling.

II. EXPERIMENT

The GaAs/GaInP double heterostructure used in this investigation was grown at NREL by MOCVD, with an active layer thickness of 0.5 μm . [3] We excite the sample with a 100 mW, 635 nm laser (CNI model # MDL-NS-635, specified fall time less than 3 ns) operated at 250 KHz. The laser is focused to a spot size of $1 \times 10^{-4} \text{ cm}^2$. We use a bandpass filter (Thorlabs FBH850-40) to remove scattered laser light and detect the transient PL with an Excelitas photon counting module, which produces a 2.2V 10 ns pulse for each photon detected. The PL signal is averaged (maximum 512 traces) on a 500 MHz Tektronix DPO3054 oscilloscope. To minimize detector saturation, the maximum average signal is held to $\sim 100 \text{ mV}$ using neutral density (ND) filters. In addition, to avoid clipping the detection pulses, the 8-bit vertical scale on the scope can be set no smaller than 200 mV/division, which limits the sensitivity of the signal measurement. We use ND filters to conduct the experiment at 5 different laser powers spanning 2 orders of magnitude in photoexcitation density. An example of the measured PL signal along with the measured system response (using a Thorlabs FBH650-40 bandpass filter to collect scattered laser light) is shown in Fig. 1.

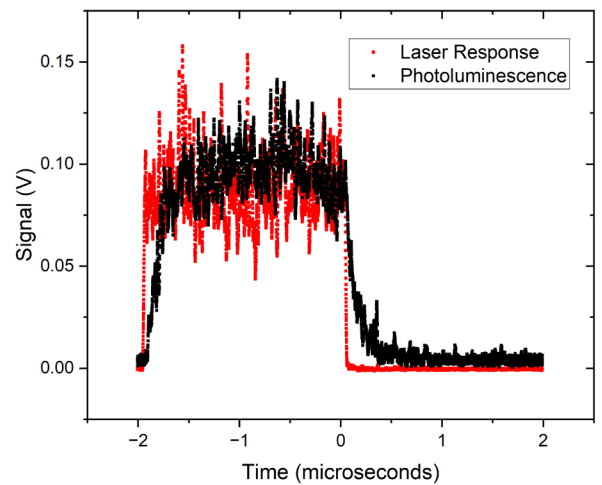


Fig. 1. Transient PL signal under low laser excitation (approximately 10 W/cm² in the ON state) showing the slow rise and fall relative to the system response (laser measurement in red).

III. RESULTS

The carrier density calibration method described in Refs. 1 and 2 involves a series of numerical steps. First, we determine the quasi-steady state generation rate G_{QSS} (electron-hole pairs per cm^3) using the measured laser power (corrected for reflections and duty cycle), photon energy, and excitation volume. These values are used to obtain a relationship between the quasi-steady-state PL signal and generation rate. Similarly, we use the measured laser signal to obtain the non-quasi-steady-state generation rate G_{NQSS} as a function of time during the transient PL measurement. Next, we use the measured transient PL signal to obtain G_{QSS} as a function of time and solve for the density-dependent recombination rate:

$$\frac{d\Delta n}{dt} = G_{NQSS} - G_{QSS} \quad (2)$$

Numerically integrating this result yields the nonequilibrium carrier density Δn as a function of time. Finally, using Equation 1, we can solve for the lifetime as a function of carrier density. The results of this analysis are presented in Fig. 2.

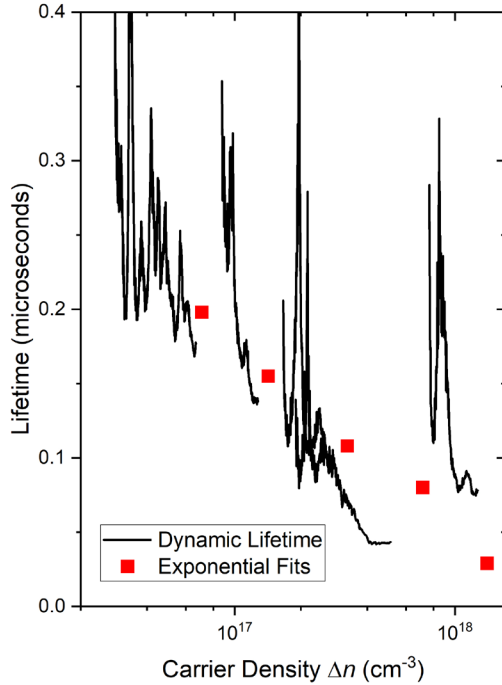


Fig. 2. Dynamic lifetime vs. photoexcited carrier density in GaAs. Also shown in red are lifetimes deduced from exponential fits to the PL decay, plotted at the initial quasi-steady-state carrier density.

The noise in the dynamic lifetime results can be attributed to the experimental limitations described in Section II. When we fit the PL decay curves to single exponentials, we obtain the lifetimes shown in red on Fig. 2. Since the carrier density changes during the decay process, we plot these measurements at the quasi-steady state density that we deduce from our dynamic calibration, immediately prior to switching the laser off. Hence, these points represent traditional lifetime estimates

informed by the dynamic calibration of the photoexcited carrier density. The measurements show a more consistent downward trend in lifetime with increasing carrier density, as expected for radiative recombination. [4]

These preliminary results can be compared with our prior analysis of the steady-state photoluminescence intensity from the same structure (sample #WA540). [5] In the power density regime studied here (approximately $10 - 1000 \text{ W/cm}^2$), that analysis indicated majority carrier densities near 10^{17} cm^{-3} and minority carrier densities ranging from 10^{13} to 10^{17} cm^{-3} . Our dynamic densities are consistent with the predicted majority densities. When we compare our measured lifetimes, which are on the order of 10^{-7} s , with the predicted radiative recombination rates ($\sim 10^8 \text{ s}^{-1}$), we observe an order of magnitude disagreement. More work will be required to fully interpret our transient results in the context of the prior model.

IV. CONCLUSION AND FUTURE WORK

Our primary conclusion is that the experimental method is viable for characterizing recombination in GaAs, but a detector that produces lower voltage pulses and/or an oscilloscope with higher vertical resolution and averaging will yield cleaner results. Alternatively, the Excelitas module should be used with a traditional single photon counting system, where a time to amplitude converter (TAC) is used to generate a transient PL histogram. [6] Our future work will focus on switching to a photomultiplier tube (PMT) detector where the pulse height can be controlled by the accelerating voltage and perhaps utilizing a higher performance digital oscilloscope.

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