Understanding the short time dynamics of transient photoluminescence experiments

Guillaume Vidon[1], Stefania Cacovich[1,3], Marie Legrand[1,2], Daniel Ory[1,2], Daniel Suchet[1,3], Jean-Baptiste Puel[1,2] , Jean-François Guillemoles[1,3]

1 IPVF, Institut Photovoltaïque d'Ile-de-France, 30 RD 128, 91120 PALAISEAU, France
2. EDF R&D, 30 RD 128, 91120 PALAISEAU, France
3 CNRS, Ecole Polytechnique, Institut Photovoltaïque d'Ile-de-France UMR 9006, 30 RD 128, 91120 PALAISEAU, France

Time resolved photoluminescence (TR-PL) is a widely used contactless tool for material characterization[1]. However, interpreting decays observed on emerging materials for photovoltaics such as perovskite absorbers is challenging. For instance, disentangling bulk from interface recombination is far from obvious[2]. Drift-diffusion models have allowed for material parameter fitting, but their complexity implies that multiple sets of parameters can correctly fit one experimental curve. Studies where the laser excitation power is varied are a common practice, reducing the uncertainty on the fitted parameters by checking that a single model can explain both high and low injection regimes. In this study, we used drift-diffusion models to interpret short time dynamics of the PL decay. We found that most of the information gained when performing a power study is contained in the short time dynamics. By a theoretical analysis of the drift-diffusion models, we established a relationship between the derivative of the PL decay just after the pulse and different model parameters such as external radiative recombination rate, absorption coefficient, diffusion coefficient along with interface and bulk recombination properties. This relationship forms a scaling law linking material and excitation parameters such as the wavelength and the fluence of the laser pulse. We first tested the validity of this scaling law via simulations and experiments with stable III-V materials. Then, we applied this method on triple-cation perovskite thin films to measure their external radiative recombination rate. This technique could bring new arguments in the ongoing debate on the precise determination of the optoelectronic properties of perovskite absorbers. The scaling law we established is a new way of interpreting TR-PL measurements, which allows us to understand the influence of material parameters on the experimental curve and to estimate them in a more direct way.

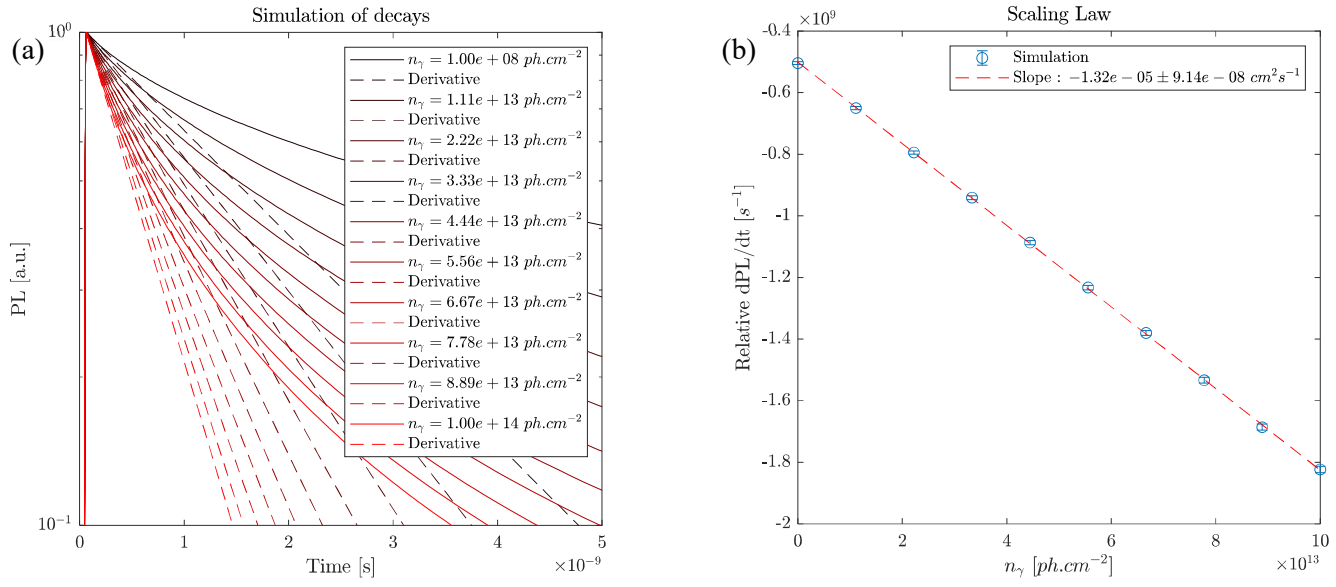

Figure 1: (a) Simulation of TR-PL decays for layers that model typical perovskite behavior. Each color corresponds to a fluence of the laser, and is composed of two curves: the simulated decay [solid line] and the fit of the beginning of the decay to determine the derivative [dotted line]. (b) Relative derivative at short time as a function of laser fluence determined for the curves of (a) [dots] and linear fitting [dotted line]. From the derived scaling law and the parameters set in the simulation, we expect a linear behavior with a slope of $-(4/3)k_2\alpha \approx -1.33 \times 10^{-5}\ cm^2/s$, with k_2 the coefficient for second order recombination $[cm^3 s^{-1}]$ and α the absorption coefficient $[cm^{-1}]$ at the laser wavelength.

1. Bercegol, A. *et al.* Slow Diffusion and Long Lifetime in Metal Halide Perovskites for Photovoltaics. *J. Phys. Chem. C* **122**, 24570–24577 (2018).

2. Weiss, T. P. *et al.* Bulk and surface recombination properties in thin film semiconductors with different surface treatments from time-resolved photoluminescence measurements. *Sci. Rep.* **9**, 5385 (2019).