Characterizing Transient and Continuous Regimes of Hot Carriers for Photovoltaics

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One of the major energy loss mechanisms in photovoltaic conversion is carrier thermalization, also known as carrier relaxation, lowering of approximately 25 percentage points the conversion efficiency [1]. Microscopically, this process involves the interaction of high-energy (hot) carriers with the surrounding lattice, resulting in a loss of excess energy as heat. A key challenge in developing solar cells capable of harvesting these hot carriers lies in understanding and characterizing the thermalization process.

Although numerous theoretical and experimental studies have investigated hot carrier thermalization, they typically focus on either the transient regime or the steady-state regime, rarely both. To the best of our knowledge, only two studies address both regimes concurrently, one theoretical [2] and one experimental [3]. In the transient regime, carriers are suddenly excited, and their relaxation is tracked over time. In contrast, the steady-state regime involves continuous carrier generation, relaxation, and recombination. These two regimes offer both overlapping and complementary insights into thermalization, yet they are often studied in isolation. Bridging this gap is essential for a more unified understanding and an efficient comparison of different characterization techniques. Moreover, most experimental works report phenomenological quantities such as the thermalization factor Q (steady-state regime) and the cooling time τ (transient regime), which are often not derived from a common, well-established thermalization model. This lack of a unified framework makes cross-regime comparison difficult.

In this study, we first establish a theoretical link between the quantities Q and τ. We find that their relationship depends on the carrier density. This dependence becomes particularly important when the cooling time is not negligible compared to the carrier decay time, making direct comparison less straightforward. We then justify the existence and relevance of these phenomenological parameters through modelling and numerical simulations of thermalization in the simplest case: a bulk polar semiconductor. This confirms the theoretical relationship previously derived. Photoluminescence data are currently being analyzed to test and extend the proposed model to more complex materials such as III-V multiple quantum wells and perovskites.

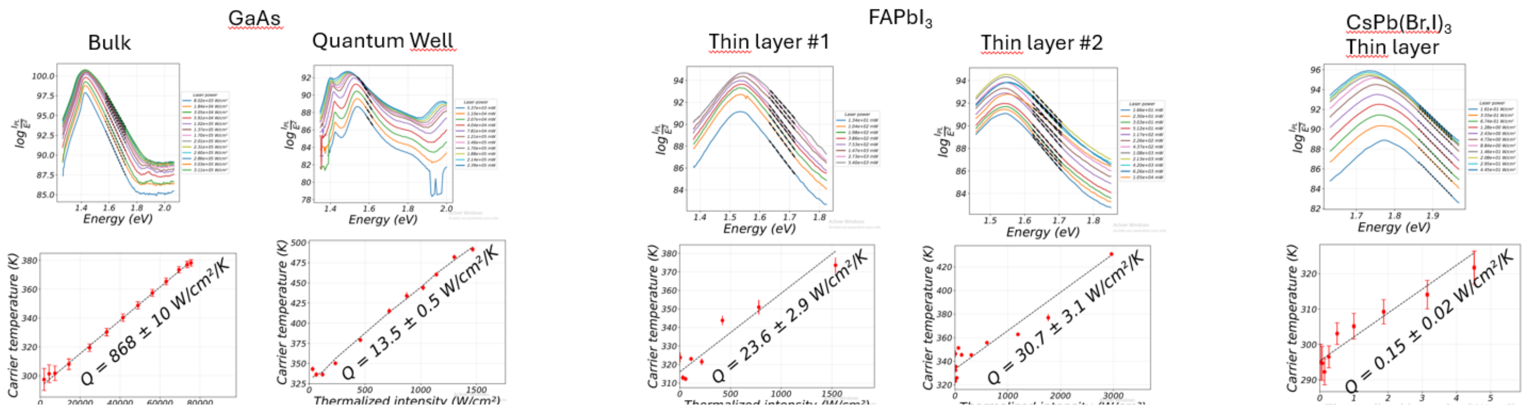

Figure 1 : Continuous-wave photoluminescence spectra of a few III-V and perovskite samples. Their thermalization factors Q are reported.

[1] Guillemoles, J.-F.; Kirchartz, T.; Cahen, D.; Rau, U. Guide for the Perplexed to the Shockley–Queisser Model for Solar Cells. *Nat. Photonics* **2019**, *13* (8), 501–505. https://doi.org/10.1038/s41566-019-0479-2.

[2] Kumekov, E.; Perel', V. I. Energy Relaxation of the Electron-Phonon System of a Semiconductor under Static and Dynamic Conditions.

[3] Fang, H.-H.; Adjokatse, S.; Shao, S.; Even, J.; Loi, M. A. Long-Lived Hot-Carrier Light Emission and Large Blue Shift in Formamidinium Tin Triiodide Perovskites. *Nat Commun* **2018**, *9* (1), 243. https://doi.org/10.1038/s41467-017-02684-w.