Two-photon excitation has seen increasing attention as a robust method for separately investigating carrier dynamics in the bulk and at the surfaces of semiconductor materials and devices. Barnard et al. used time-resolved photoluminescence (PL) to separate bulk and surface lifetime components in CdTe crystals [1], whilst Yamada et al. used this to study the carrier diffusivity and photon recycling effect for perovskite crystals [2], [3]. In two-photon microscopy, a sub-band gap femtosecond-pulsed laser is focused into a semiconductor sample using a high numerical (NA) aperture lens. This produces localized carrier generation at the focal point due to the non-linear two-photon absorption effect. The resultant carrier dynamics and PL signal reflect the carrier dynamics near the excitation focal point. Hence, by varying the position of the excitation focal point, the bulk dynamics are probed with reduced influence from the surfaces. However, a comprehensive description of the carrier dynamics and PL is currently non-existent, with previous studies making broad assumptions regarding the generation rate and excess carrier profiles, such as the use of bi-exponential decays, which are generally invalid for semiconductor materials [1], [3]–[5]. Here, for the first time, we present a comprehensive mathematical model for the carrier dynamics and PL signal in a semiconductor material due to two-photon absorption. The state-of-the-art model considers both transient and quasi-steady state excitation methods and allows use of injection-dependent parameters such as the effective carrier lifetime and the diffusivity. We apply this model to a slab of uniformly-doped silicon to assess the potential and limitations of two-photon excitation for extracting surface and bulk recombination parameters of silicon. It was found that due to the long diffusion length in silicon, it is challenging to extract the bulk lifetime if the surface recombination is the dominant recombination mechanism. Full results will be discussed in the full paper.

Figure 1: Initial excess carrier profiles for a 200 µm thick p-type Si wafer doped at $10^{15}$ cm$^{-3}$. The inset shows the excellent localization of the carrier profile near the focus for NA = 1.4.

Figure 2: PL signal as a function of focus depth using the quasi-steady-state excitation method (generation rate based on NA = 1.4 from Figure 1) for various surface-recombination velocities.

We developed the first instance of a mathematical model for two-photon excitation in semiconductor materials, and applied this model to the ubiquitous semiconductor silicon. Importantly, this model is readily adaptable to any other semiconductor material. Furthermore, it is expected that this framework can be extended to other existing characterization methods which probe the excess carrier concentration, including light-beam induced current and photo-conductance.