Above-bandgap non-linear transmission of Si reveals a two-photon absorption edge at 1.7 eV

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Silicon plays a key role in modern society as it forms the basis of our digital infrastructure. However, relatively little is known about the non-linear absorption of silicon at wavelengths below 850 nm with, to the best of our knowledge, only one previous study from 1990 [1]. We aim to contribute to the field by describing non-linear absorption of silicon from 600 nm to 1100 nm. By combining thorough experimental characterization and advanced modelling, we are able to distinguish between two-photon absorption (2PA) and three-photon absorption (3PA) coefficients.

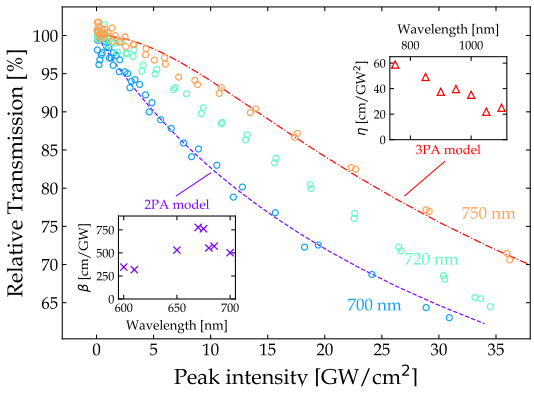

Figure 1: Non-linear transmission curves for silicon at wavelength 750, 720, and 700 nm demonstrating the change in curve shape around 730 nm. The inserts show fitted 3PA (η) and 2PA (β) coefficients at wavelengths 600-1100 nm.

The non-linear transmission curves are acquired using the z-scan method [2] with a wavelength-tunable fs-second laser on a 4 µm thick freestanding silicon thin-film. Detailed temporal and spatial characterization of the laser pulses allows us to obtain non-linear transmission as a function of peak intensity. With this, we can go beyond the standard linear approximation often made when analyzing open-aperture z-scans (eq. (12) in [2]) which critically fails to determine the order (i.e. 2PA or 3PA) of the non-linear process under study.

The relative-transmission curves clearly separate into two distinct shapes depending on the spectral regime as shown in Fig. 1. For wavelengths below 730 nm, the relative transmission has a concave dependence on peak intensity, while above 730 nm the curves are flatter and convex at low peak intensities. The observed transition wavelength at 730 nm, i.e. a photon energy of 1.7 eV, is at half the direct band-gap energy of silicon, and thus the 2PA edge which supports the interpretation that non-linear absorption below/above 730 nm is dominated by 3PA/2PA, respectively. To capture this behavior, we use exact theoretical 2PA and 3PA models in their respective regimes to determine absorption coefficients shown in the inserts of Fig. 1. The 2PA coefficients show a dependence on photon energy similar to a recent theoretical study [3], and in line with this study we find that the 2PA coefficients have previously been significantly underestimated. Furthermore, we present 3PA coefficients for wavelengths between 600-1100 nm, which to the best of our knowledge have not previously been reported.

References:

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