A theoretical study of multi-photon absorption using the Z-scan technique

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We report on the theoretical analysis of two-, three-, four- and five- photon absorption processes using the open aperture Z-scan technique. The simulation for two-photon absorption was performed with a theoretical model present in the literature. For three-, four- and five-photon absorption we proposed a set of new equations, which can be used in the fittings of the experimental data. The input parameters were chosen based on those typical for organic materials. The simulations showed that for the process in which a large number of photons are simultaneously absorbed, a narrowing in the shape of the Z-scan curves can be observed, due to the need of higher pump irradiance. We also simulated measurements of the normalized transmittance change as a function of the incident irradiance, and we could observe distinct slopes for curves, which make our simulations adequate for the study of the multi-photon absorption in organic compounds.

Nonlinear absorption coefficient

$$\alpha(I) = \alpha + \alpha_2 I + \alpha_3 I^2 + \alpha_4 I^3 + \ldots$$

Change in irradiance

$$\frac{dI}{dz} = -\alpha_n I^n$$

Generic solution

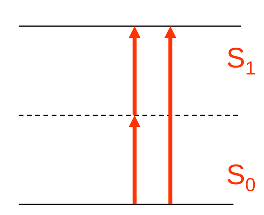
$$I(z,r,t) = \frac{I_0}{\sqrt[n-1]{1+(n-1)\alpha_n L I_0^{(n-1)}}}$$

Non-linear transmittance for the process of MPA

2PA

$$T(L) = \sum_{m=0}^{\infty} \frac{[-q_0]^m}{(m+1)^{3/2}}$$

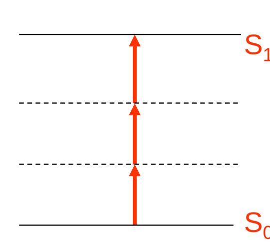
$$q(0) = \frac{I_0 \alpha_2 L \omega_0^2}{\omega_z^2}$$



3PA

$$T(L) = \frac{1}{\sqrt{\pi}\sqrt[3]{2\alpha_3 L I^2}} \int_0^1 \frac{R(x)}{x\sqrt{-\ln x}} dx$$

$$R(x) = \ln\left(\sqrt{1+2\alpha_3 L I^2 x^2} + \sqrt{2\alpha_3 L I^2 x^2}\right)$$

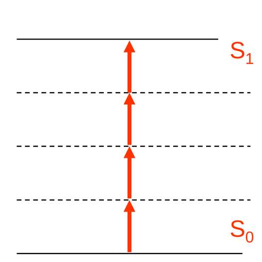


4PA

$$T(L) = \frac{1}{3\sqrt{\pi}\sqrt[3]{3\alpha_4 L I^3}} \int_0^1 \frac{R(\Delta)}{x\sqrt{-\ln x}} dx$$

$$R(\Delta) = \ln\left(\frac{\sqrt{\Delta(x)^2 + \Delta(x) + 1}}{\Delta(x) - 1}\right) - \sqrt{3}\arctan\left[\frac{2\Delta(x)+1}{\sqrt{3}}\right] + \frac{\sqrt{3}\pi}{2}$$

$$\Delta(x) = \sqrt[3]{1+\left(3\alpha_4 L I^3 x^3\right)^{-1}}$$

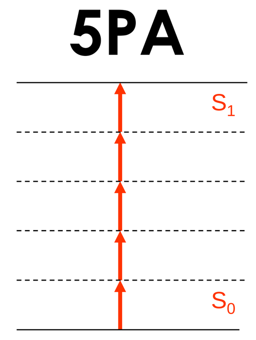


5PA

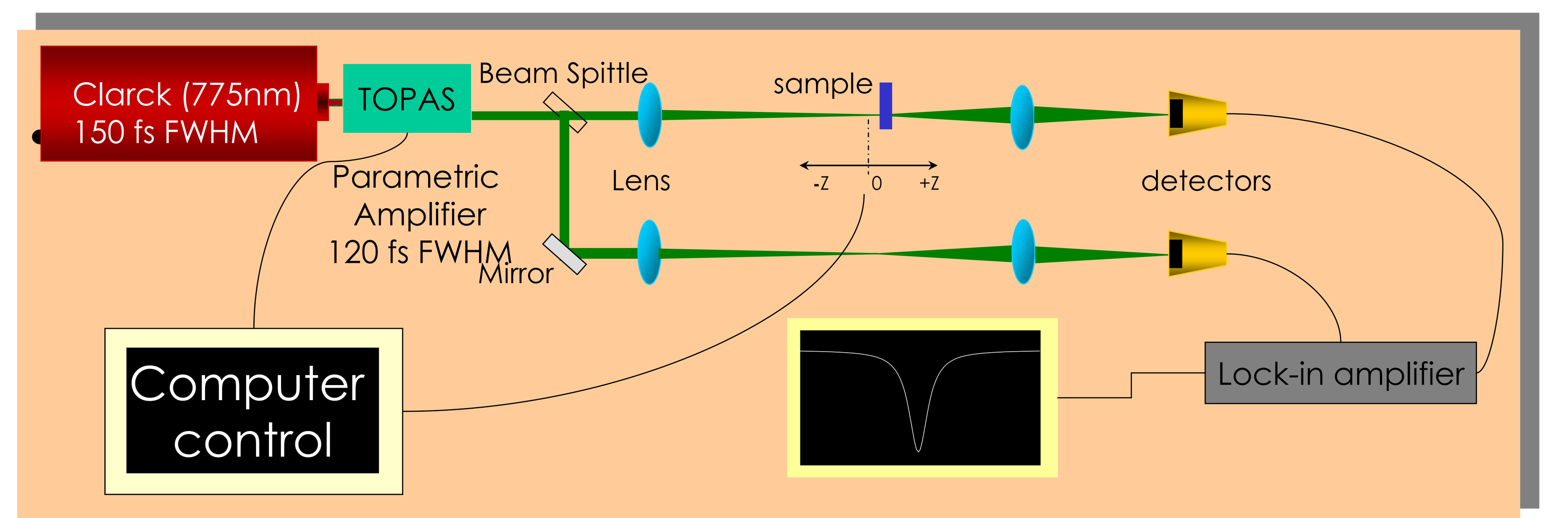
$$T(L) = \frac{1}{2\sqrt{\pi}\sqrt[4]{4\alpha_5 L I^4}} \int_0^1 \frac{R(\Delta)}{x\sqrt{-\ln x}} dx$$

$$R(\Delta) = \ln\sqrt{\frac{\Delta(x)+1}{\Delta(x)-1}} - \sqrt{3}\arctan[\Delta(x)] + \frac{\pi}{2}$$

$$\Delta(x) = \sqrt[4]{1+\left(4\alpha_5 L I^4 x^4\right)^{-1}}$$



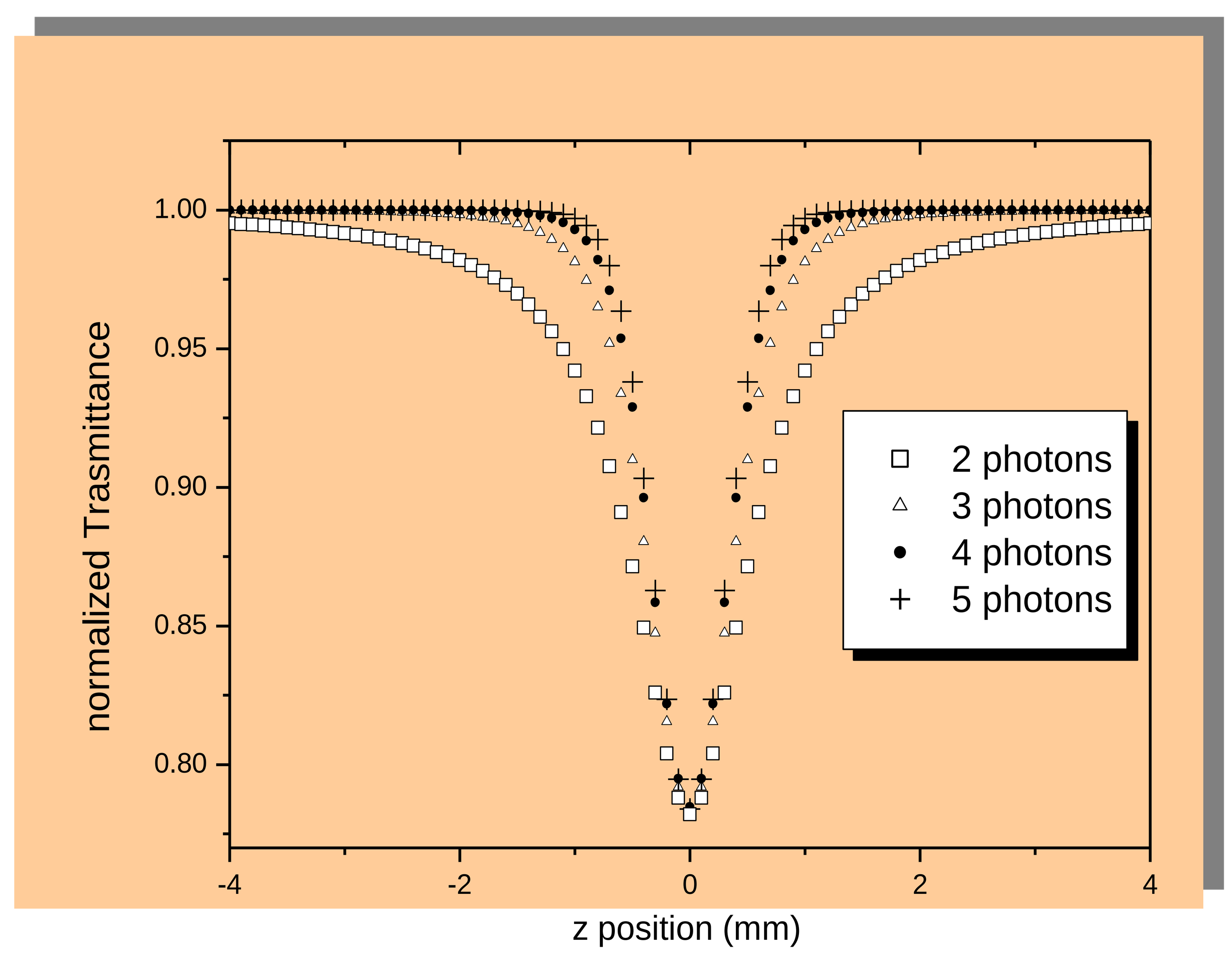
✓ Z-Scan experimental setup



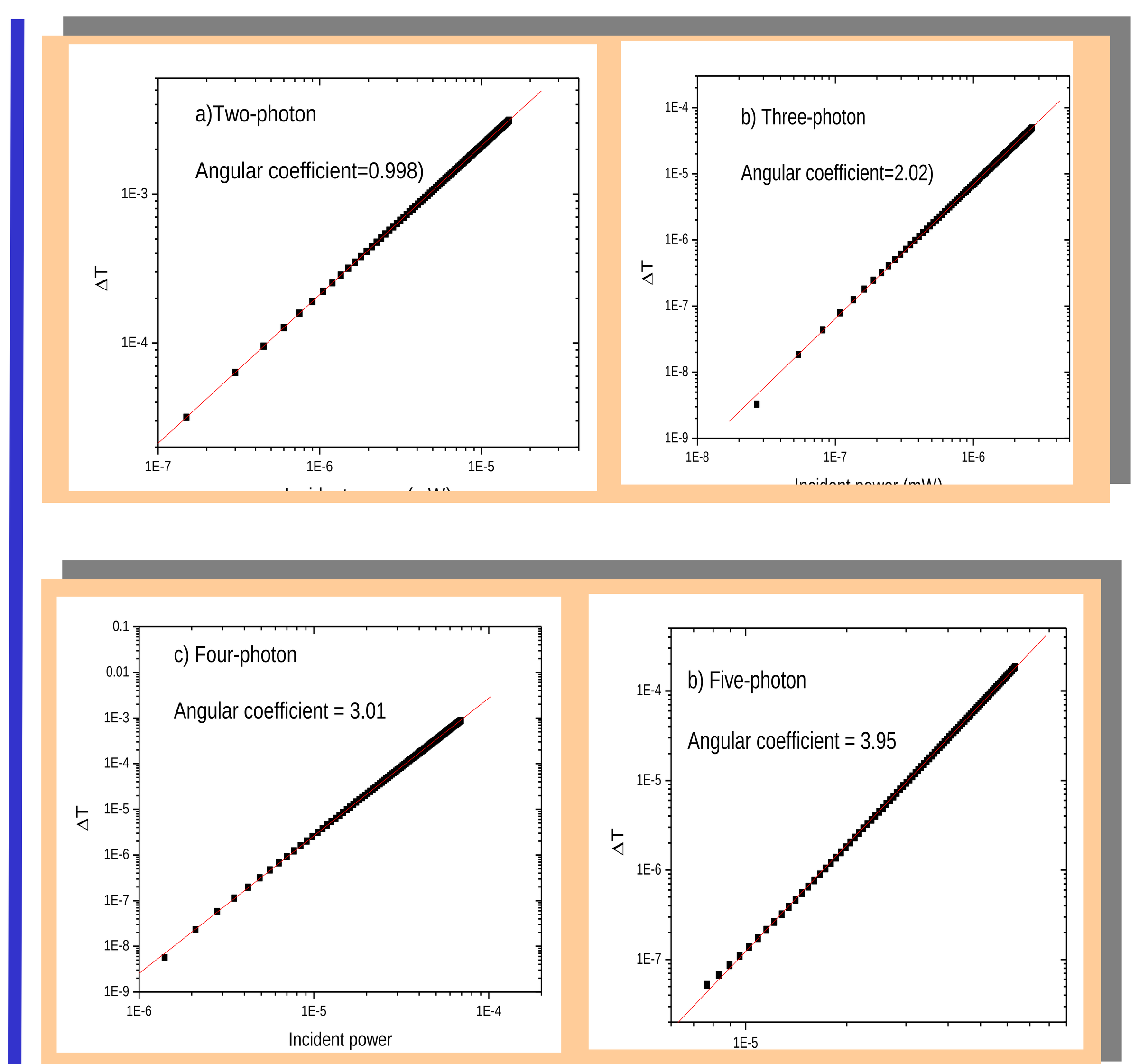
Input parameters:

- **λ = 500 nm until 1600 nm**
- **w_0 = 14 µm until 18 µm**
- **τ_{pulso} = 150 fs**
- **F = 1KHz**

Distinct shapes for the Z-scan curves for 2-, 3-, 4- and 5-photon absorption



Narrowing of the Z-scan curves for higher values of n due the need of a higher pump intensity



The simulated curves for normalized transmittance change (ΔT) as a function of the excitation beam showed different slopes for the two-, three-, four-, and five-photon absorption processes .

✓ Conclusions

Through theoretical simulations employing the Z-scan technique, we have demonstrated the validity of this method to study the process of multi-photon absorption (2PA, 3PA, 4PA, and 5PA) in organic materials. The simulations of the normalized transmittance as a function of the incident irradiance provided distinct slopes for the curves. The two-, three-, four- and five-photon absorption cross-section values obtained with the simulations were typical for organic compounds.

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