Saturation effects in nonlinear absorption and refraction of DO_3 (Disperse Orange-3) Solution.

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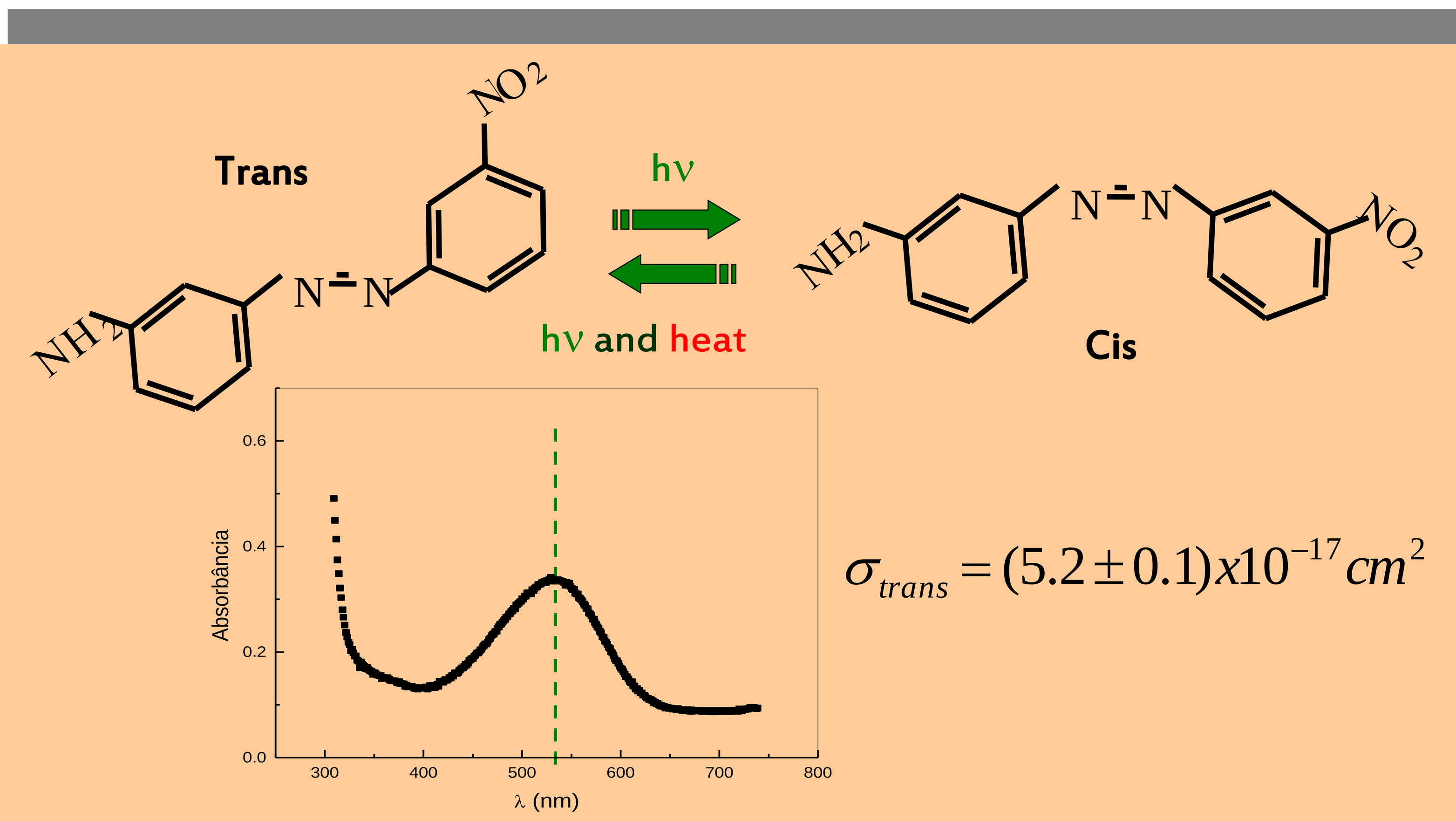
Abstract

The Z-scan technique has been used to study the saturation of both absorption and nonlinear refraction (n_2) in a DO3 solution. We observed that the saturation of absorption is due to the of photoinduced cis conformation. A three-energy-level model was applied to describe this effect and the absorption cross-section was obtained. Besides, a saturation of n_2 was observed. Further investigations must be done to determine the origin of this process.

The DO_3 molecule (azobenzene-compound)

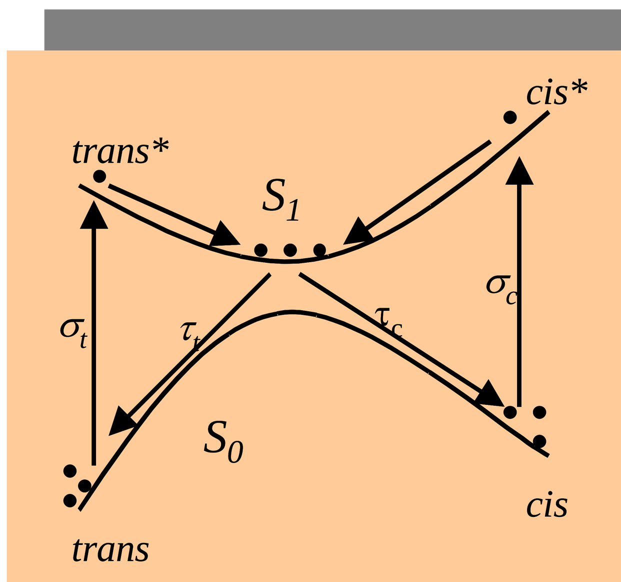
✓ Photoisomerization process



$$\sigma_{trans} = (5.2 \pm 0.1) x10^{-17} cm^2$$

Azobenzene undergoes *cis-trans* photochemical isomerization after excitation to the $S_1(n\pi^*)$ state band. The insert graphic shows the absortion of DO_3 in DMSO solvent.

✓ Three-energy-level diagram model and rate equations



$$\frac{dn_{trans}}{dt} = -n_{trans}W_{trans} + \frac{n_{S1}}{\tau_{trans}}$$

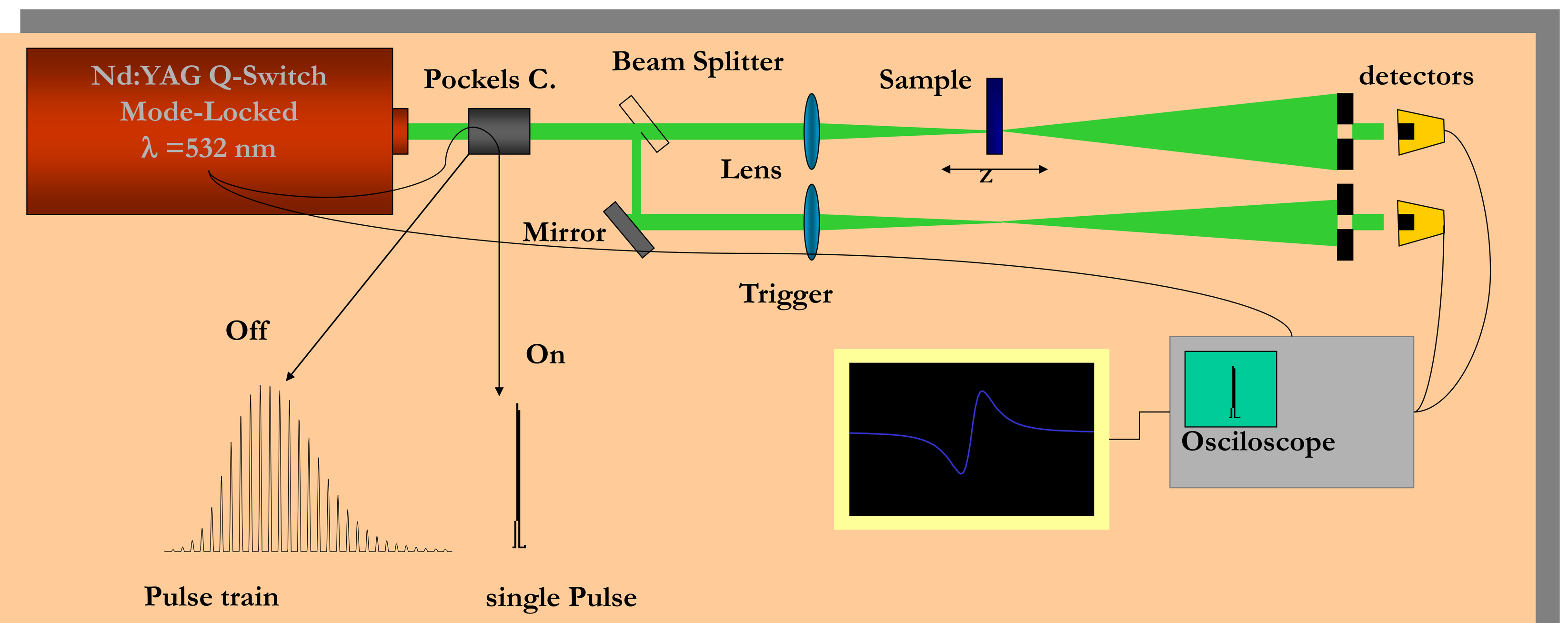
$$\frac{dn_{S1}}{dt} = n_{trans}W_{trans} + n_{cis}W_{cis} - \frac{n_{S1}}{\tau_{trans}} - \frac{n_{S1}}{\tau_{cis}}$$

$$\frac{dn_{cis}}{dt} = -n_{cis}W_{cis} + \frac{n_{S1}}{\tau_{cis}}$$

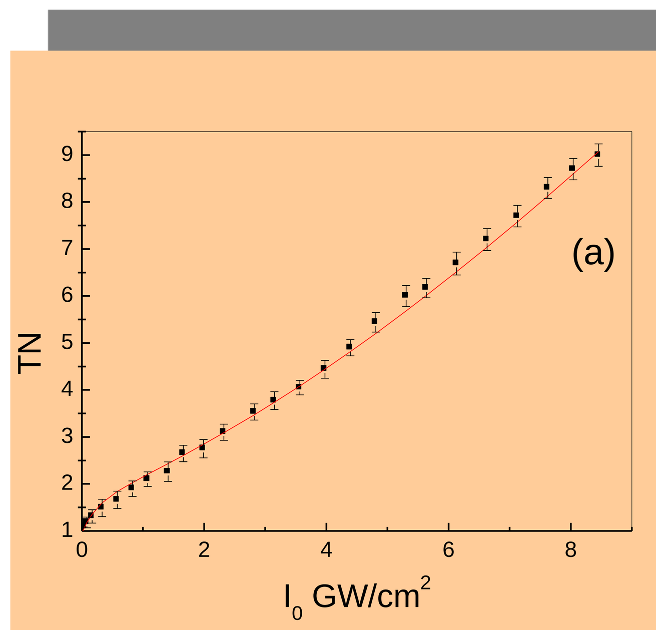
$$\alpha(t) = N\{n_{trans}\sigma_{trans} + n_{cis}\sigma_{cis} + n_{S_1}\sigma_{S_1}\}$$

The results obtained with the Z-scan technique with open and closed aperture (presented in Fig. (a) and (b)) can be described by the three-energy-level. Azobenzene molecules, initially in the S_0 (*trans*) band, are excited to the S_1 (*trans**) band, relaxing nonradiatively to the bottom of this band. At this point, there are two possible relaxation pathways: one to S_0 (*trans*) and other to S_0 (*cis*) state, both with similar relaxation life times (σ_t and σ_c). This process transfers part of the population from S_0 (*trans*) to S_0 (*cis*). An analogous mechanism happens with molecules in S_0 (*cis*). However, in this case the process is less efficient because the *cis* cross-section is smaller than that of the *trans* absorption. After several photoisomerization cycles, a population of molecules in the S_0 (*cis*) ground state band is created, which generates the saturable nonlinear absorption and refraction.

✓ Z-Scan experimental setup



Our Z-scan experiment used single pulses extracted from the pulse train of a frequency-doubled Q-switched and mode-locked Nd:YAG laser, operating at 532 nm with a10 Hz repetition rate. The FWHM pulse duration was 100 ps, and the spatial profile of the laser beam was approximately Gaussian. The intensity of laser was varied using a calcite polarizer.



Parameters fitted

nonlinear absorption

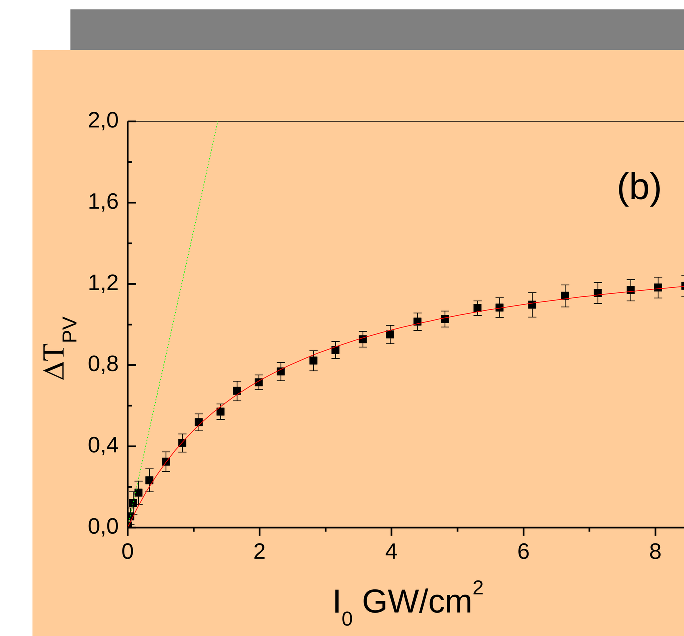
τ_t, τ_c = (2.3K0.4) ps

$$\sigma_{cis} = (4.2 \pm 0.1) x10^{-17} cm^2$$

nonlinear refraction

n_2 = -1.1 x10^{-12} cm^2/W

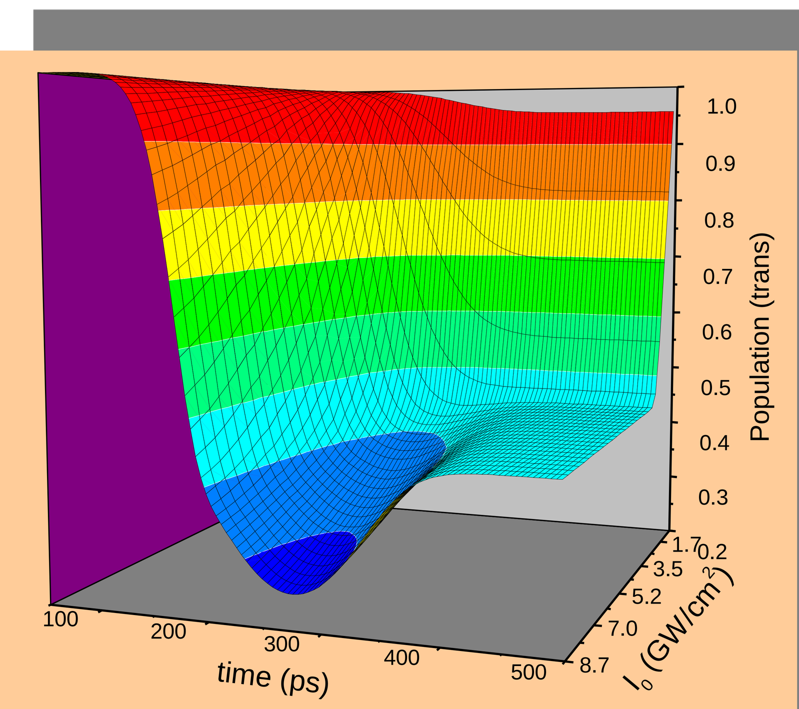
n_{2eff} = -0.5 x10^{-12} cm^2/W

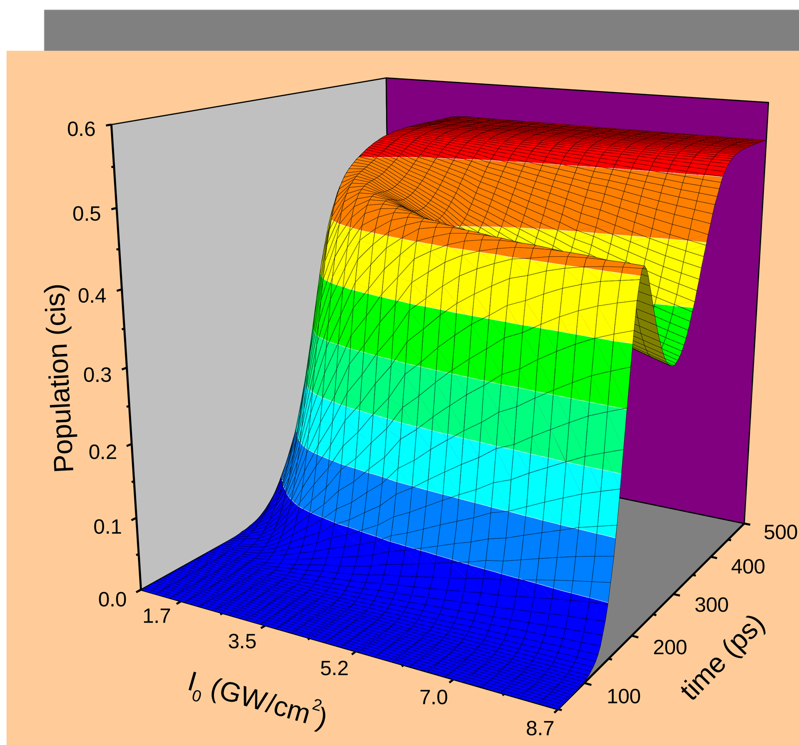


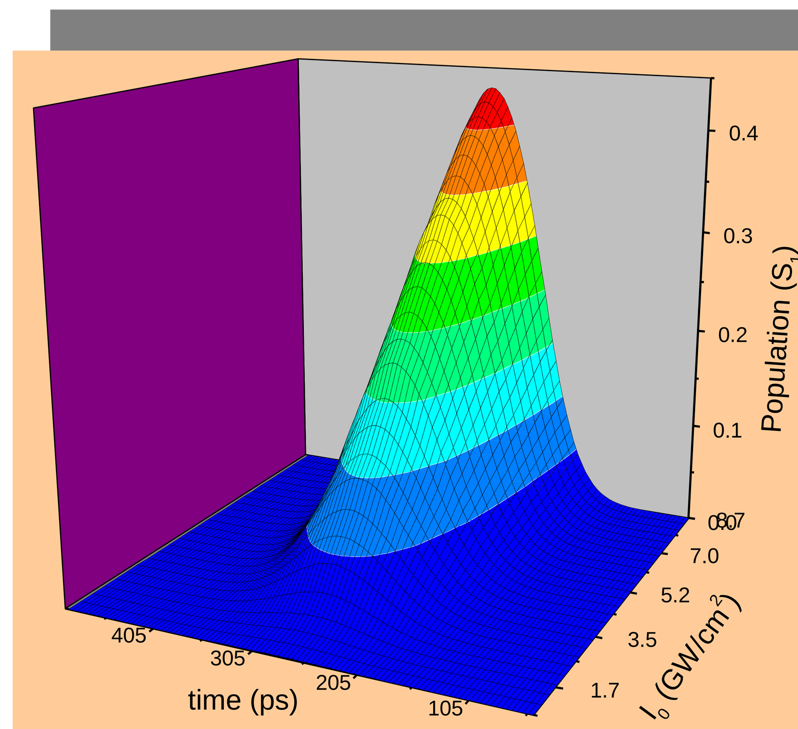
$$\Delta n = n_{2eff} I / (1 + I / I_s) \qquad (1)$$

Figure (a) Normalized transmittance (TN) as function of intensity for open aperture Z-scan measurements. The solid line is the fitting obtained using the three-energy-level diagram model. (b) The effect of saturation in close aperture Z-scan measurements. The solid line represents the fitting obtained with the eq. (1). The dashed line represents the nonlinear refraction effect without saturation.

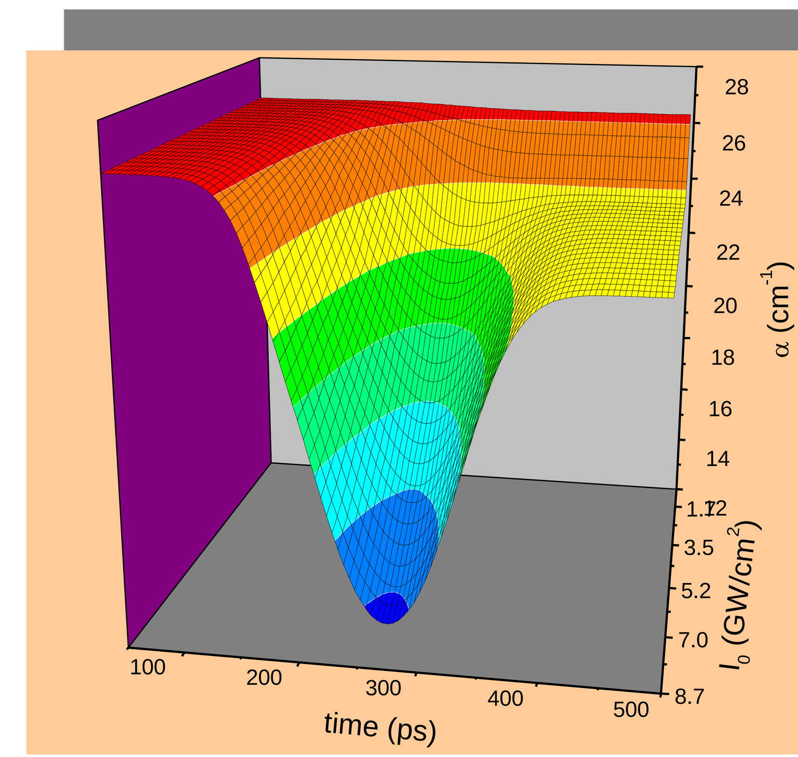
✓ Intra-pulse dynamic population







✓ Intra-pulse absorption coefficient



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