Optimization of the two-photon excited fluorescence via pulse shaping in Y-shaped molecules

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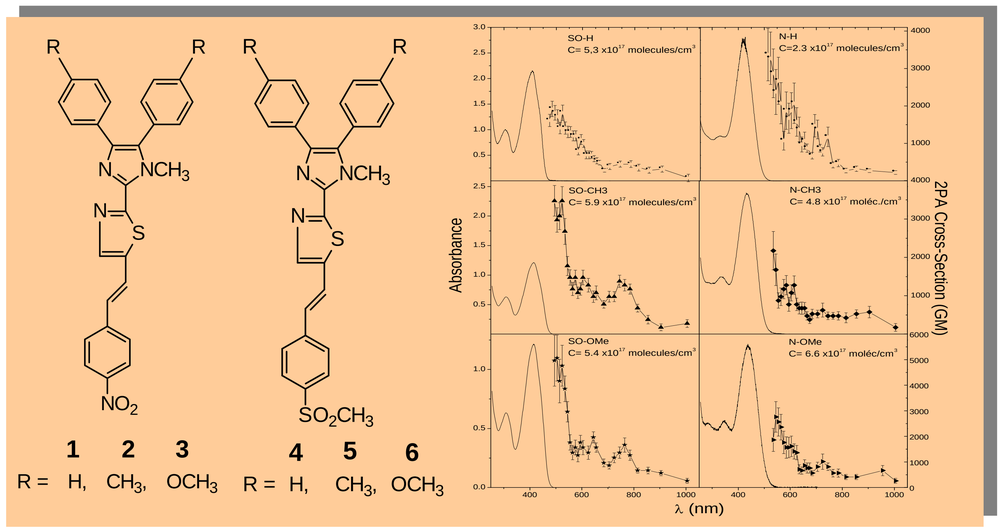
Abstract

In this study we report the two-photon excited fluorescence (2PEF) and its optimization via pulse shaping in a novel classes of chromophores, named as Y-shaped compounds, which presents large two-photon absorption (2PA) cross-section. The 2PEF for all molecules could be optimized using a deformable mirror (DFM) and a genetic algorithm (GA}. A typical increase of five times was observed in the 2PEF for the best solution, comparing with the flat DFM. In order to check the two-photon nature of the observed process, 2PEF measurements were carried out for different pump beam powers. The quadratic dependence on the excitation power observed confirms the pure two-photon process, as expected. These compounds are very promising for use in two-photon fluorescence microscopy.

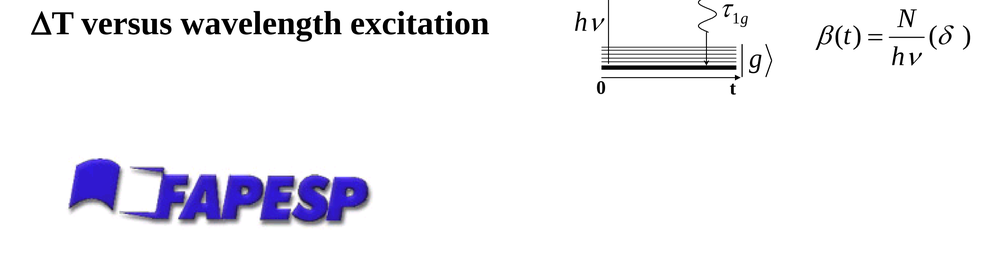
"Y-shaped" chromophores.

✓ Molecular structures, absorbance and 2PA spectra.

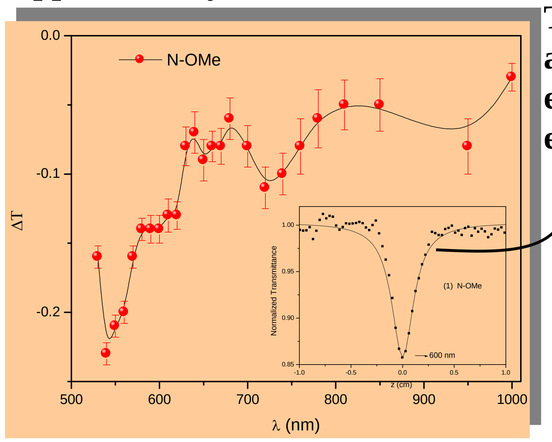


The 2PA spectra are obtained with the Z-scan technique

✓ Z-Scan experimental setup and typical results



Our Z-scan experiment employ laser pulses from a commercial optical parametric amplifier (TOPAS) pumped by a 150 fs pulses at 775 nm delivered by a Ti:sapphire chirped pulse amplified system (CPA-2001, from Clark-MXR Inc.), operating at 1kHz repetition rate. The FWHM pulse duration from TOPAS was about 120 fs, and the spatial profile of the laser beam presented an approximately Gaussian distribution.

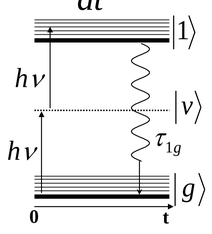


ΔT versus wavelength excitation

Typical curve of 2PA. The fit are obtained by rate equations that describe two–energy-level diagram.

$$\frac{dn_g(t)}{dt} = -n_g(t)W_{g1} + \frac{n_1(t)}{\tau_{1g}}$$

$$\frac{dn_1(t)}{dt} = +n_0(t)W_{g1} - \frac{n_1(t)}{\tau_{1g}}$$

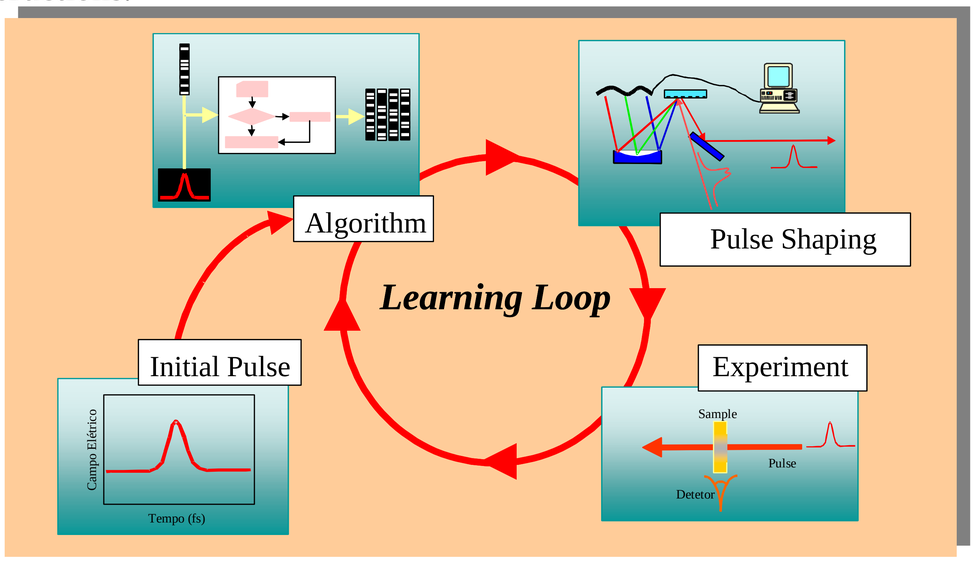


$$W_{g1} = \frac{\delta_{g1} I^2}{2h\nu}$$

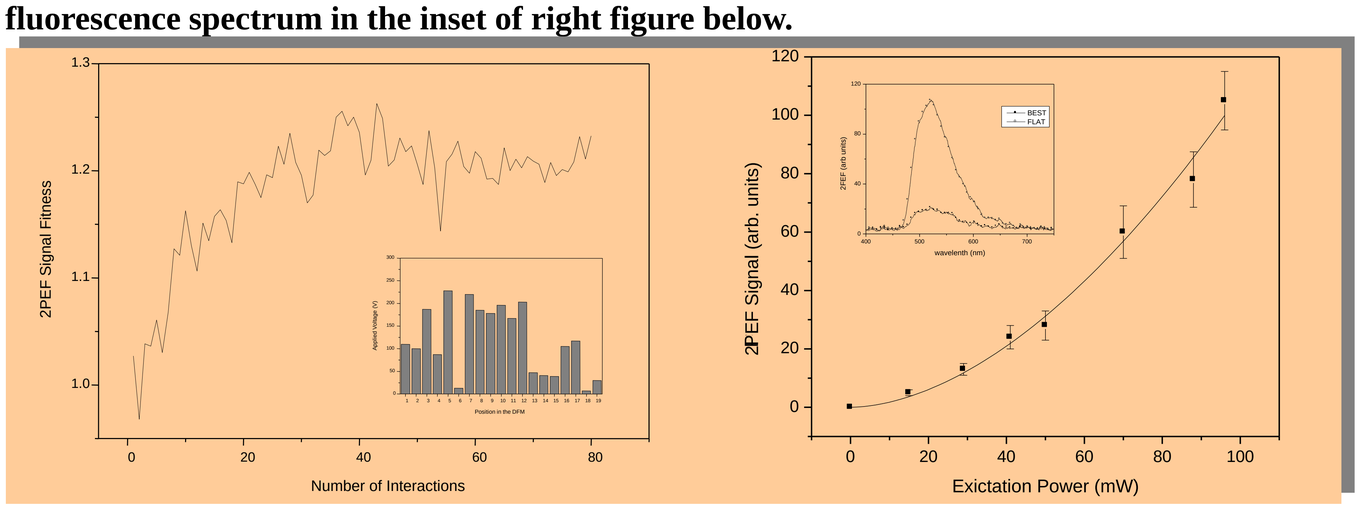
$$\beta(t) = \frac{N}{h\nu}(\delta)$$

✓ Coherent control of the 2PEF

The experimental set-up for measuring the two-photon excited fluorescence employed femtosecond pulses from a Kerr-Lens-Modelocked (KLM) Ti:sapphire oscillator ~ 800 nm. The fluorescence spectrum was collected with a conventional fiber optic spectrophotometer. A micro-machined deformable mirror (DFM) was used to shape the spectral phase in a zero-dispersion stretcher configuration. Optimization of the 2PEF is obtained using an evolutionary strategy which begins with a set of random pulse shapes whose associated 2PEF signal is measured. Those pulses that produce the most intense fluorescence are retained, duplicated, perturbed, and reproduced, as the GA requires. This process is repeated until a desired number of interactions.



The fitness signal evolution as a function of the number of interaction for SO-Ome is shown in the left figure below, where a clear increase in the fitness can be observed. The applied voltage in the DFM actuators for the best pulse optimized is showed in the inset. Approximately the same solutions were found for the other compounds. A typical increase of five times was observed in the 2PEF for the best solution, comparing with the flat DFM, as show the fluorescence spectrum in the inset of right figure below.



The 2PEF measurements were carried out for different pump beam powers, in order to check the two-photon nature of the observed process. The right figure above, shows that the fluorescence signal exhibits a quadratic dependence on the excitation power, as expected for pure two-photon processes. The solid line in this figure represents the theoretical fitting obtained with a quadratic polynomial function.

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