Two- and three-photon excited fluorescence in Y-shaped molecules

L. De Boni, L. Misoguti, C. R. Mendonça
Instituto de Física de São Carlos - USP - São Carlos, SP, Brazil

Abstract

In this work we have studied the two- and three-photon excited fluorescence on a new series of Y-shaped chromophores, using pulses at 750 nm and 1400 nm from an optical parametric amplifier pumped by 150 fs pulses from a Ti:sapphire chirped pulse amplified system. The measured two- and three-photon absorption cross-sections are in the order of $1000 \times 10^{-50} \text{ cm}^2$ and $5 \times 10^{-28} \text{ cm}^2$, respectively. These cross-section values are obtained with z-scan technique.

**“Y-shaped” chromophores.**

- **Molecular structures, absorbance and 2PA spectra.**
- **Z-Scan experimental setup and typical results.**

Our Z-scan experiment employ laser pulses from a commercial optical parametric amplifier (TOPAS) pumped by 150 fs pulses at 775 nm delivered by a Ti:sapphire chirped pulse amplified system (CPA-2001, from Clark-MXR Inc.), operating at 1 kHz 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.

- **Multiphoton excited fluorescence**

We have used pulses from an optical parametric amplifier (TOPAS, from Light Conversion) pumped by 150 fs pulses at 775 nm delivered by a Ti:sapphire chirped pulse amplified system (CPA-2001, from Clark-MXR Inc.) operating at 1 kHz repetition rate as the excitation source. The optical parametric amplifier was operated at 750 nm and 1400 nm in order to achieve two- and three-photon excitation, respectively, according to the linear absorption spectra of the studied samples. The full-width at half-maximum (FWHM) pulse of the TOPAS is about 120 fs. The excitation beam was focused with a lens of focal distance f=20 cm onto a 2 mm path length cell, filled with the solution under investigation. The multi-photon excited fluorescence was collected perpendicularly to the incident beam direction and measured using an Acton spectrometer (Spectra Pro 150).

The two- and three-photon nature of the observed processes was verified by measuring the multi-photon excited emission dependence on the excitation laser irradiance.

The figure in the right side shows the fluorescence spectrum for S-OH obtained for excitation at 490 nm (one-photon), 750 nm (two-photon) and 1400 nm (three-photon). The y-scale was adjusted to clarify presentation. As expected these spectra are identical, indicating that the emission corresponds to the relaxation from the first excited state for all excitation wavelengths employed. As can be seen from this figure, when excited at 750 nm, the observed emission is related to a two-photon absorption from $S_0$ to $S_2$. However, when excitation at 1400 nm is employed only three-photon absorption processes can promote molecules to $S_0$, from which relaxation occur.

In order to demonstrate the two- and three-photon nature of these excited fluorences, we have measured the emission intensity as a function of the pump beam irradiance, as shown in left a) and b) figures, for S-OH at 750 and 1400 nm, respectively. Similar results, not shown here, were obtained for the other samples. The solid lines in this figure represents the best linear fits obtained with slopes of 2.1 and 3.4, clearly demonstrating the two- and three-photon nature of the observed nonlinearities at 750 and 1400 nm, respectively.

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