Control of two-photon absorption in organic compounds by pulse shaping: spectral dependence

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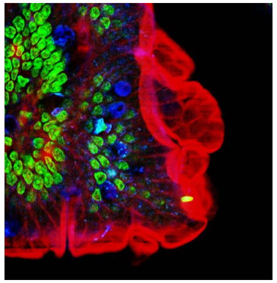
Motivation

The two-photon absorption (2PA) process has important technological applications

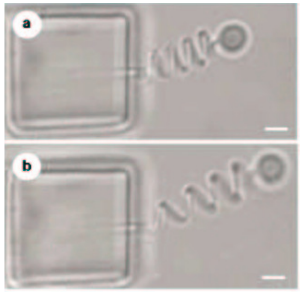
- 2PA fluorescence microscopy;

- 2PA microfabrication and photo-polymerization;

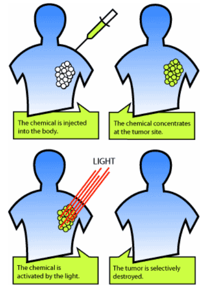
- Photodynamic therapy.



Diaspro *et al. BioMedical Engineering (2006)*

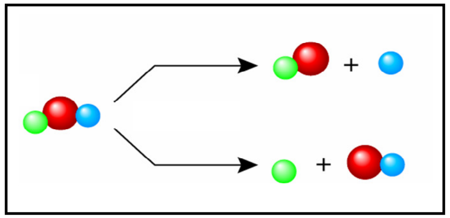


Kawata *et al*, *Nature* (2001)



Coherent control (light-matter interaction)

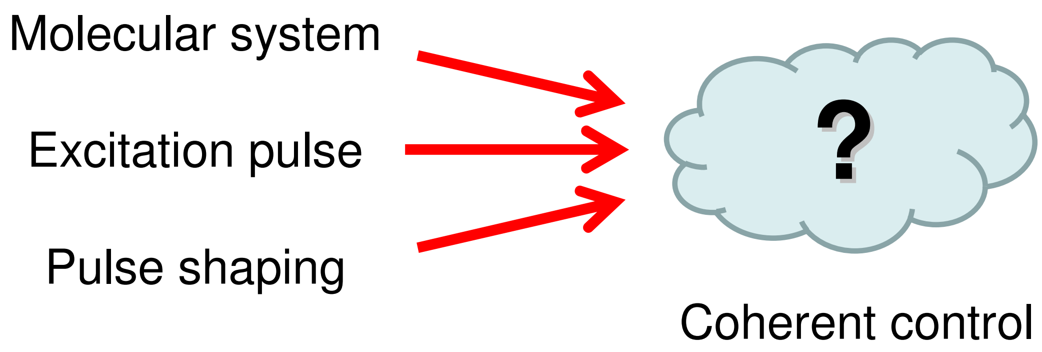
To create a particular product that is unattainable by conventional chemical or photochemical means;



Objectives:

- To obtain control schemes that could enhance (optimize) and modulate (control) interesting processes;
- To achieve a better fundamental understanding of these processes (and molecules).

Motivation



Very complex subject

Our proposal

Investigate the control of the TPA process on well-known conditions

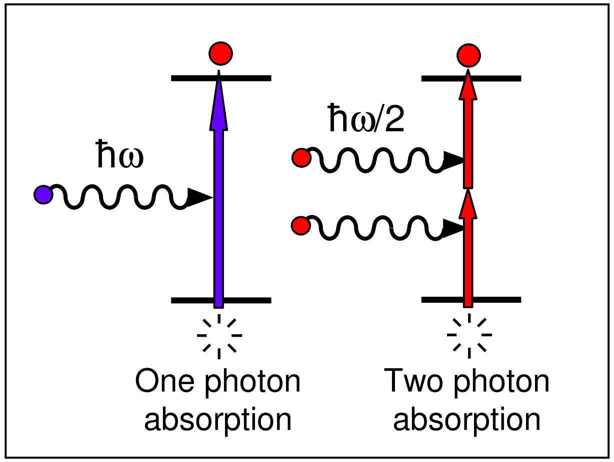
Organic compounds
Excitation pulses
Pulse shaping

Well-known features

Outline

- Control of two-photon absorption process
- Compounds studied
- Pulse shaping technique
- Results
- Summary

Control of two-photon absorption process



$$\alpha_2 \propto \mathrm{Im}\left[\chi^{(3)}\right]$$

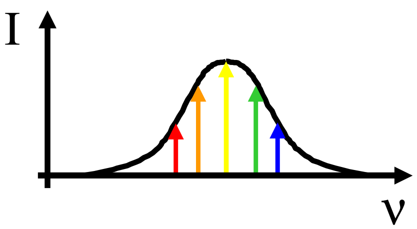
α_2 : two-photon absorption coefficient

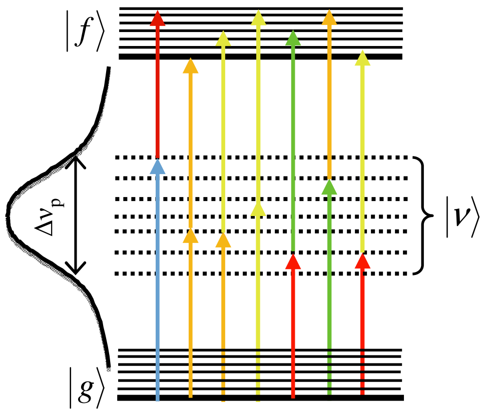
$$W_{gf}^{(2)} = \sigma_{2PA} \cdot I^2$$

W_{gf} : transition rate

High intensities ↔ Ultrashort pulses (fs)

Ultrashort pulses (fs)
↕
Spectral broadband





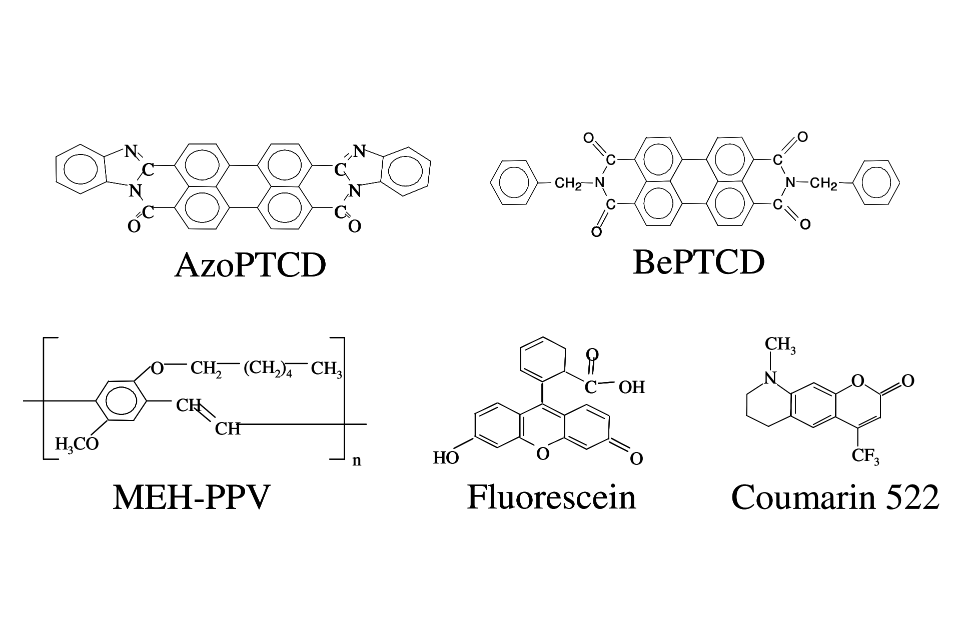
Non-degenerate 2PA process

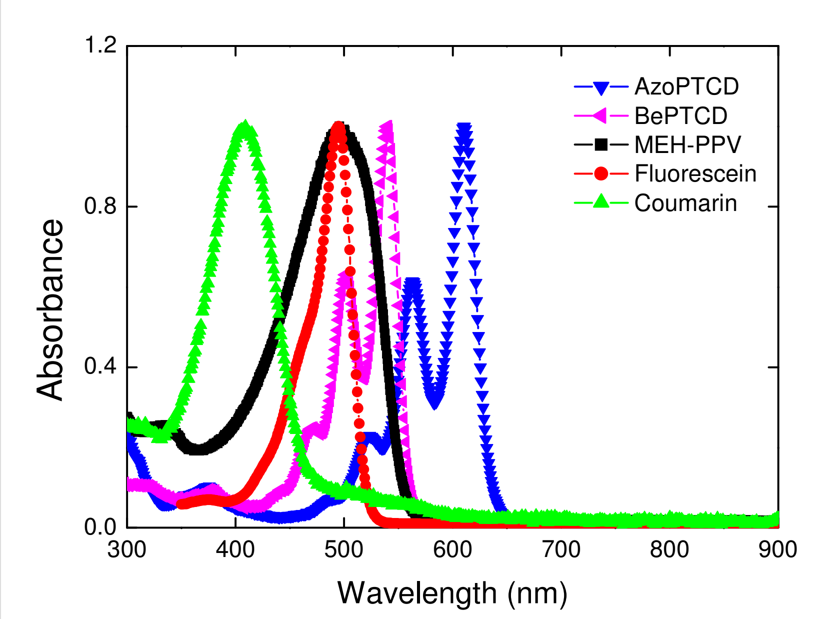
Non-degenerate 2PA

Intrapulse interference process

Coherent control

Compounds utilized

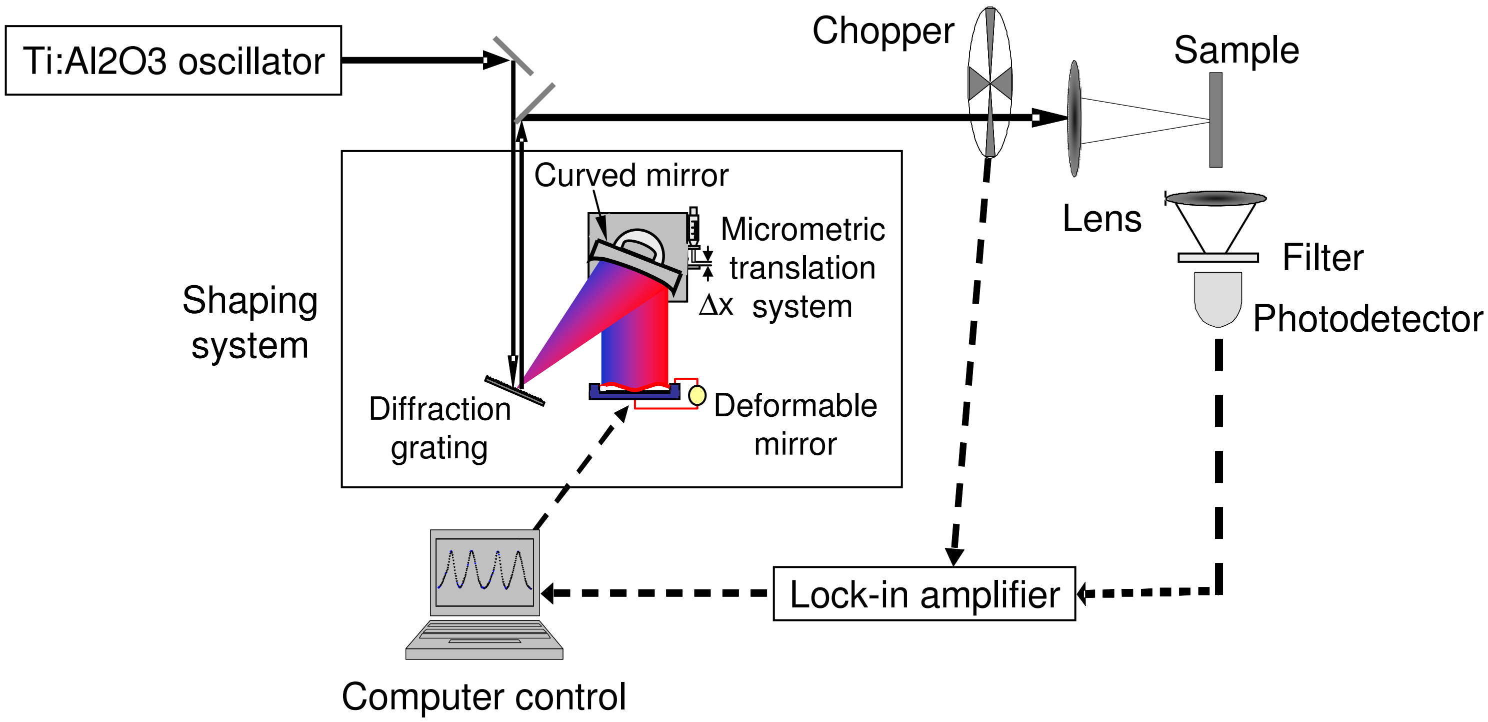




Organic compounds with specific linear absorption bands and 2PA features.

c = 0.5 mg/mL

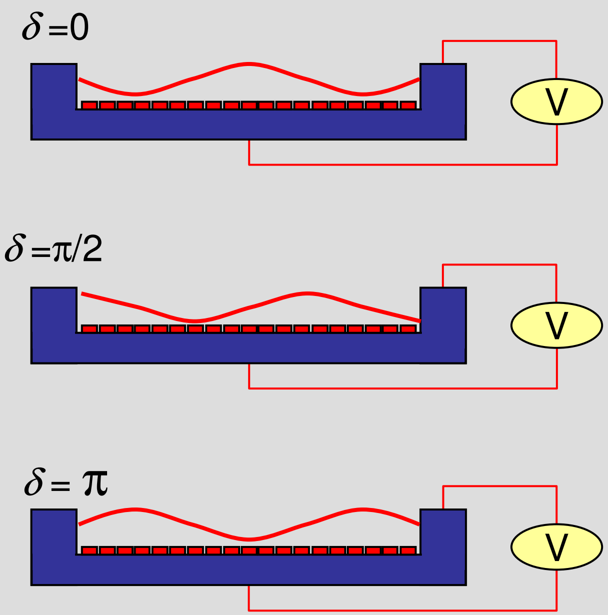
Pulse shaping technique



Pulse shaping technique

Cosine-like spectral phase modulation: $\Phi(\omega) = \alpha \cdot \cos(\beta\omega + \delta)$

$\alpha = 0.5\pi$, $\beta = 2\pi/\Delta\omega_{pulse}$;



Scan δ : 0 – 4π

Two-photon absorption process

$$S_{2PA} \propto \int_{-\infty}^{+\infty} g_{2PA}(\omega) \left| \int_{-\infty}^{+\infty} E(\omega/2 - \Omega) E(\omega/2 + \Omega) d\Omega \right|^2 d\omega$$

$$E(\omega) = A(\omega) \exp[i\Phi(\omega)]$$

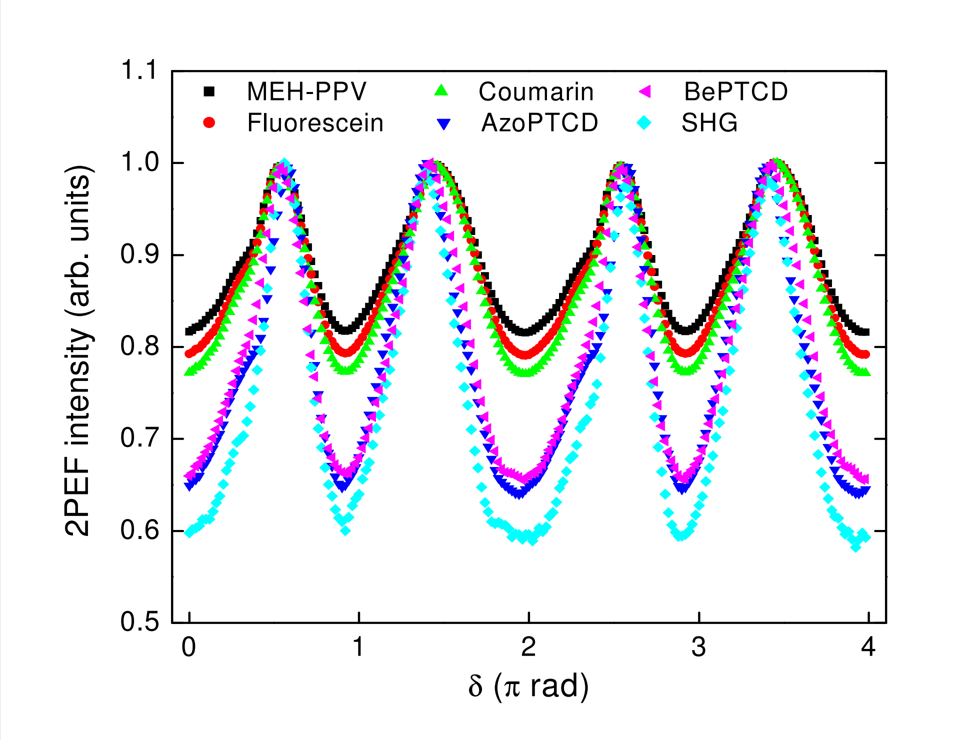
$$A(\omega/2 - \Omega) A(\omega/2 + \Omega) \cdot \exp[\Phi(\omega/2 - \Omega) + \Phi(\omega/2 + \Omega)]$$

$$\Phi(\omega/2 - \Omega) + \Phi(\omega/2 + \Omega) \begin{cases} = 0 & \text{Asymmetric phase configuration} \\ \neq 0 & \text{Symmetric phase configuration} \end{cases}$$

Results

λ_{pulse} = 800 nm,

$\Delta\lambda_{pulse}$= 40 nm (FWHM)



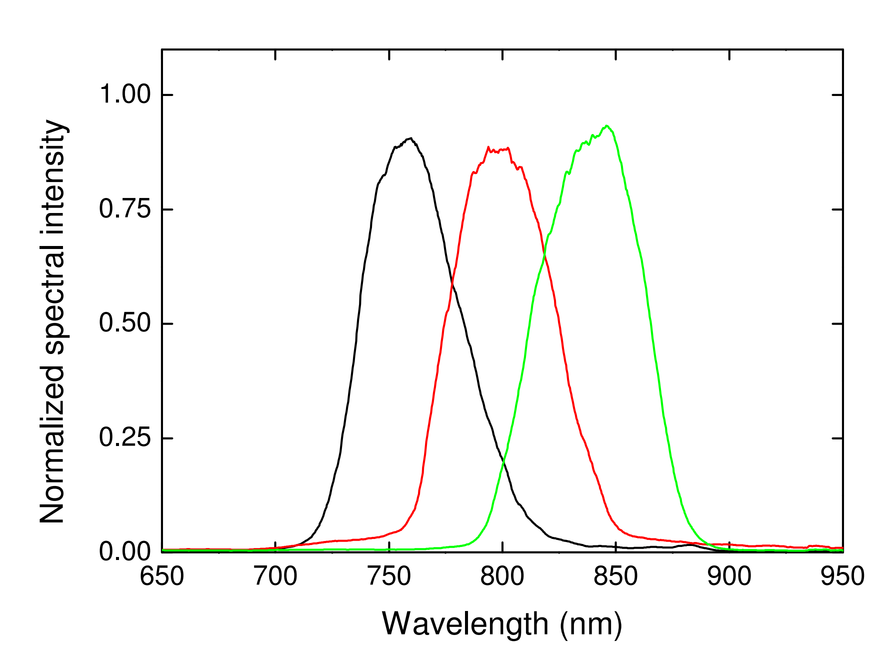
2PEF: two-photon excited fluorescence

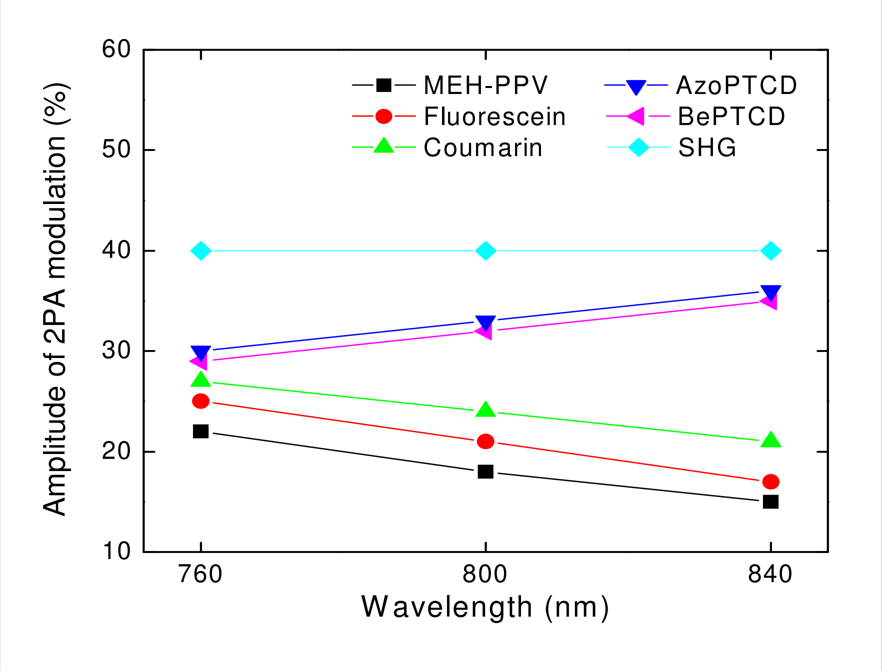
$\Phi(\omega) = \alpha \cdot cos(\beta\omega + \delta)$

Cosine-like spectral phase modulation

Results

λ_{pulse} = 760, 800 and 840 nm , $\Delta\lambda_{pulse}$= 40 nm





Results

Theoretical simulations

$$S_{2PA} \propto \int_{-\infty}^{+\infty} g_{2PA}(\omega) \left| \int_{-\infty}^{+\infty} E(\omega/2-\Omega)E(\omega/2+\Omega)d\Omega \right|^2 d\omega$$

MEH-PPV, Fluorescein, Coumarin 522: **Group 1**

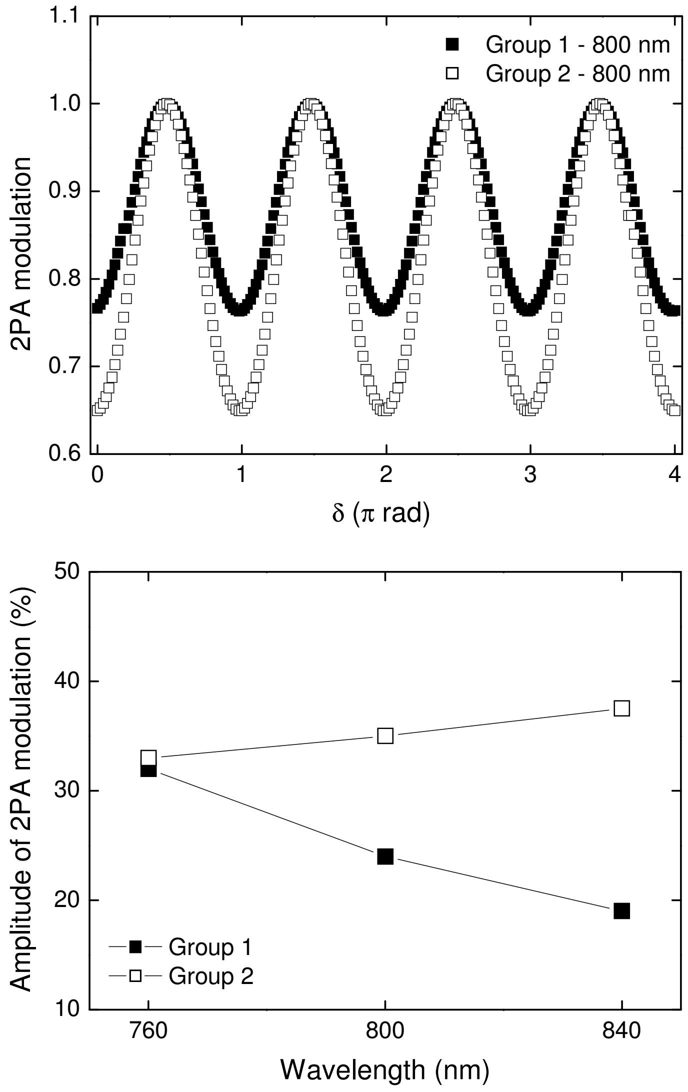
2PA peak (state) ~ 340 – 380 nm

AzoPTCD and BePTCD: **Group 2**

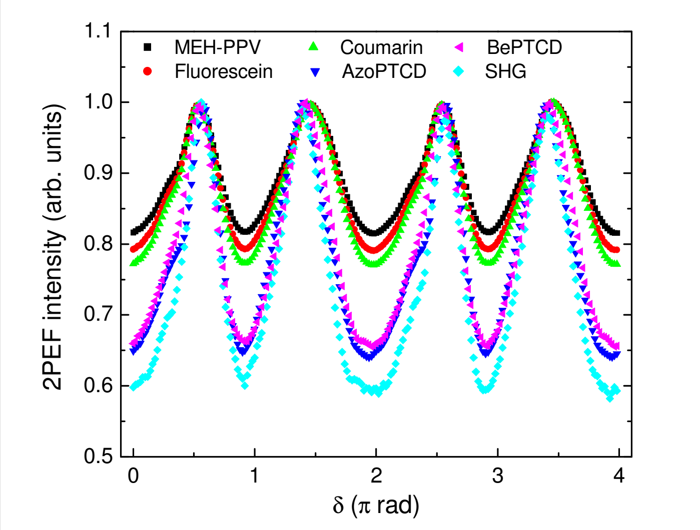
2PA peak (state) ~ 420 nm

Group 1: E_{gf} = 3.45 eV (360 nm)
Group 2: E_{gf} = 2.95 eV (420 nm)

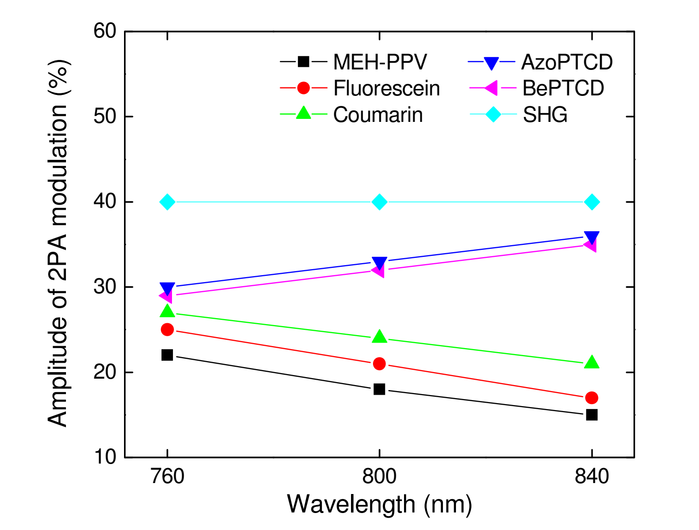
Γ_{gf} ~ 0.85 eV (80 nm)



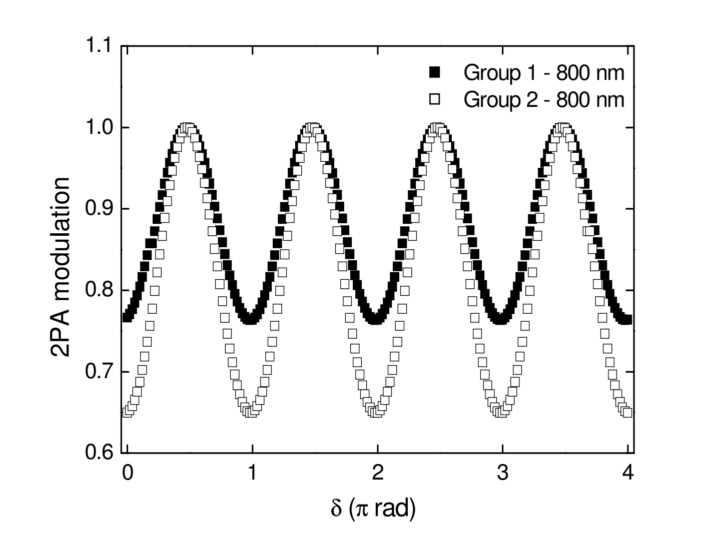
Summary



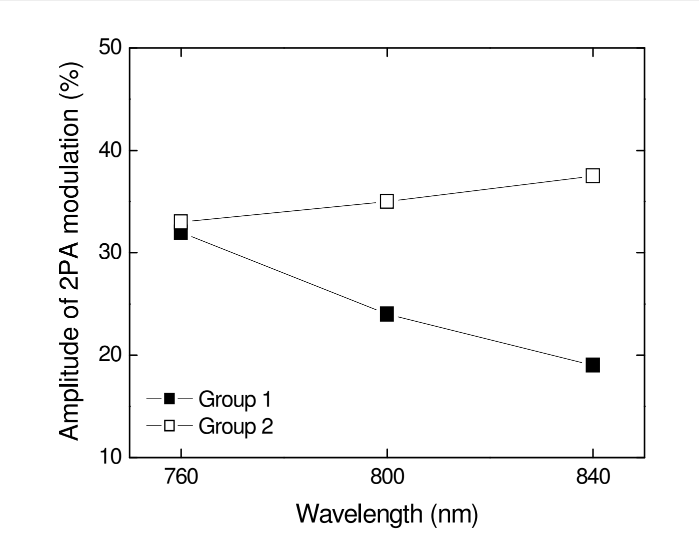
MEH-PPV
Fluorescein
Coumarin
AzoPTCD
BePTCD
SHG
2PEF intensity (arb. units)
δ (π rad)



MEH-PPV
Fluorescein
Coumarin
AzoPTCD
BePTCD
SHG
Amplitude of 2PA modulation (%)
Wavelength (nm)



Group 1 - 800 nm
Group 2 - 800 nm
2PA modulation
δ (π rad)



Group 1
Group 2
Amplitude of 2PA modulation (%)
Wavelength (nm)

Acknowledgments

FAPESP

CAPES

CNPq

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Thanks for your attention