Entangled two-photon absorption on Organic Molecules

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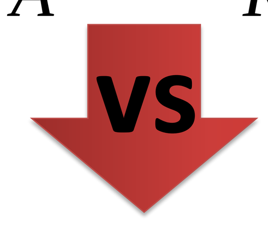
Abstract

We present the realization of an experimental setup which allow us to observe entangled two-photon absorption process occurring on organic chromophores in solution, particularly on Rhodamine B (RhB) and *meso*-Tetraphenylporphyrin (TPP) at 810 nm . We observe that by using entangled photon pairs, it is possible to induce a two photon transition on the studied molecular systems on the low photon flux regime. The setup and the studied organic chomophores may have relevant applications on new spectroscopic techniques, microscopy and photon detection among others.

Motivation

- Conventional two-photon absorption (TPA) techniques
  Require high photon flux densities ($\phi \approx 10^{20}$ photons/cm²s), high power are usually necessary.
  Random TPA cross sections (δ_R) are typically around 10^{-50} cm⁴s/photon.
- Entangled TPA (ETPA)
  A linear dependence on ϕ for the ETPA rate has been theorized and experimentally observed.[1,2]
  Low values for $\phi \approx 10^{12}$ photons/cm²s can induce the ETPA process.
  ETPA cross sections (σ_E) are around 10^{-17} cm²/molecule.[3]
- Since low ϕ values are needed when entangled photons are used as an excitation source, it may have important implications reducing the probability of photodestruction for a sample, compared with conventional TPA techniques

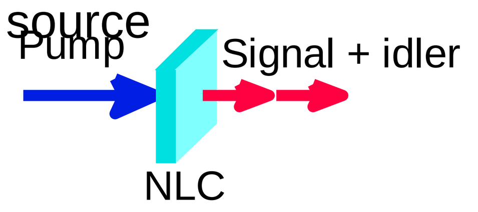
$$R_{RTPA} = \delta_R \phi^2$$



$$R_{ETPA} = \sigma_E \phi$$

Experimental Setup

Spontaneous parametric down-conversion (SPDC) is a convenient entangled photons source



$$\overrightarrow{k_p} = \overrightarrow{k_s} + \overrightarrow{k_i}$$
$$\omega_p = \omega_s + \omega_i$$

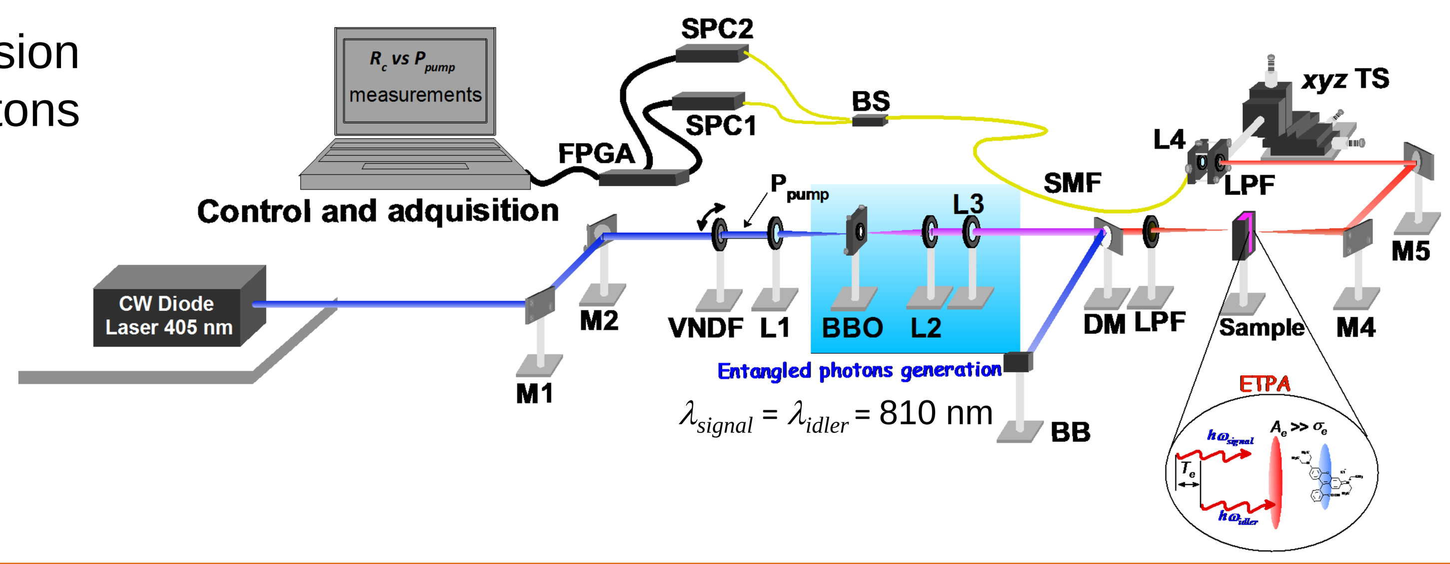


Figure 1. Experimental setup. M=mirror, L=lens, BBO=beta Barium Borate crystal, LPF=long pass filter, MP=micrometric positioner, BS=beam splitter, SPD=single photon detector and CCC=coincidence count circuit

Theoretical Model

Detected photon pairs rate without sample
(≡ SPDC photons production rate)

$$R_{c,0} = \alpha P_{pump}$$

Two-photon absorption rate

$$R_{c,blank} - R_{c,sample} = 2\,\sigma_e c\, l\, N_A \alpha_i P_{pump}$$

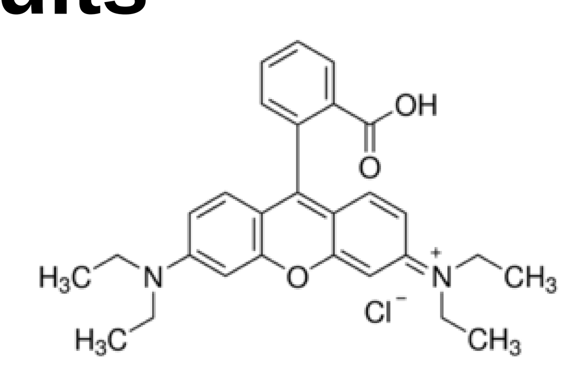
Detected photon pairs rate for a blank; Detected photon pairs rate for a sample; β = $2\,\sigma_e c\, l\, N_A \alpha$

c: sample concentration
l: cuvette path lenght
N_A: Avogadro's number

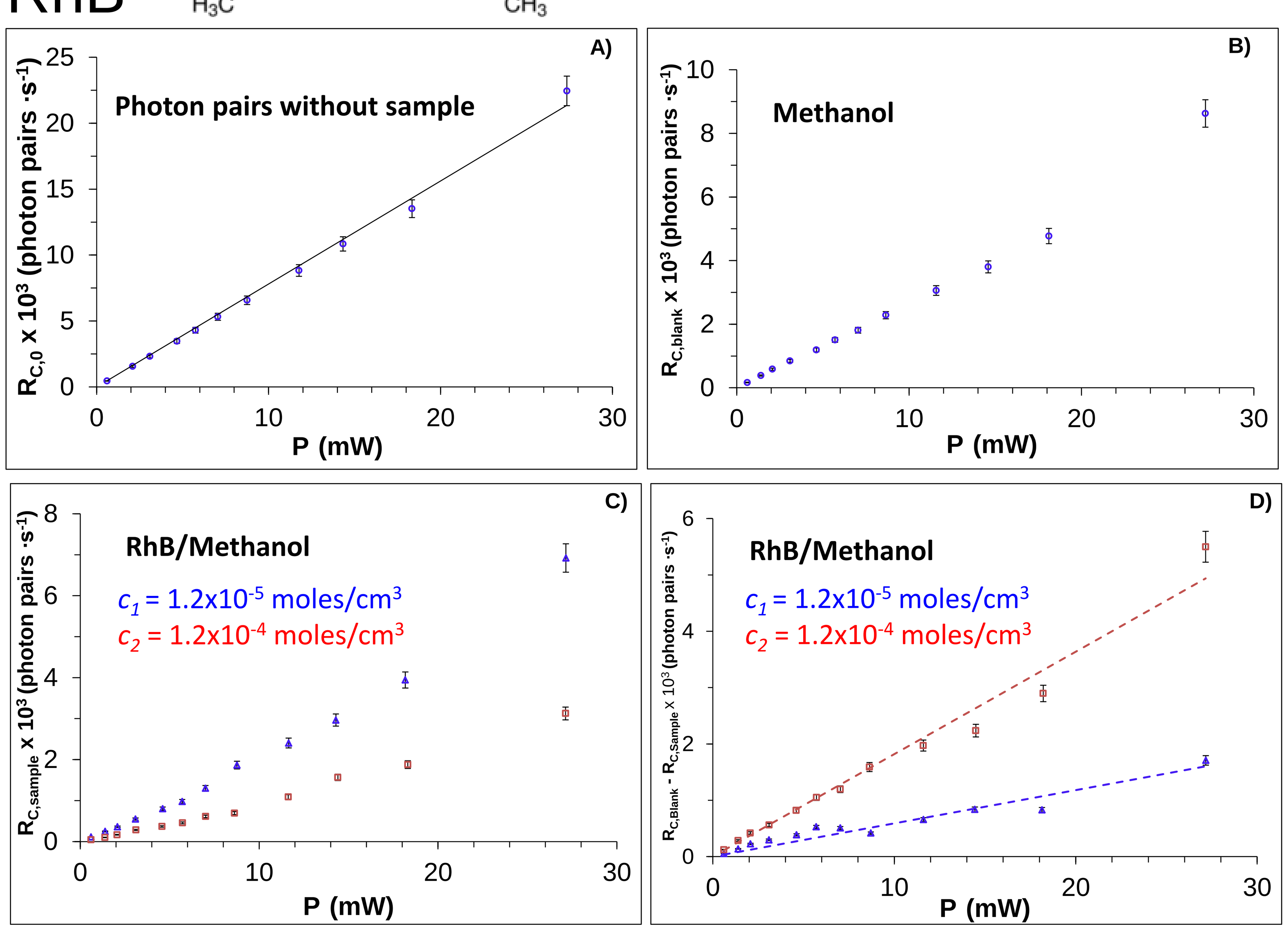
σ_E estimation

$$\sigma_E = \frac{\beta}{2c\, l\, N_A \alpha}$$

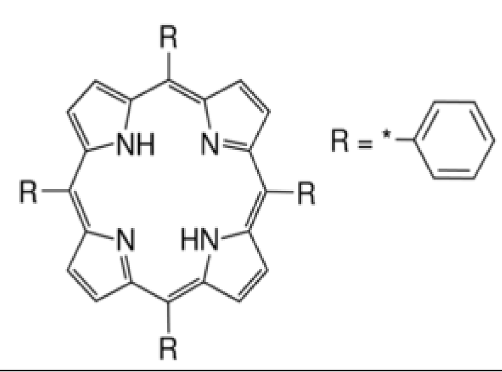
Results



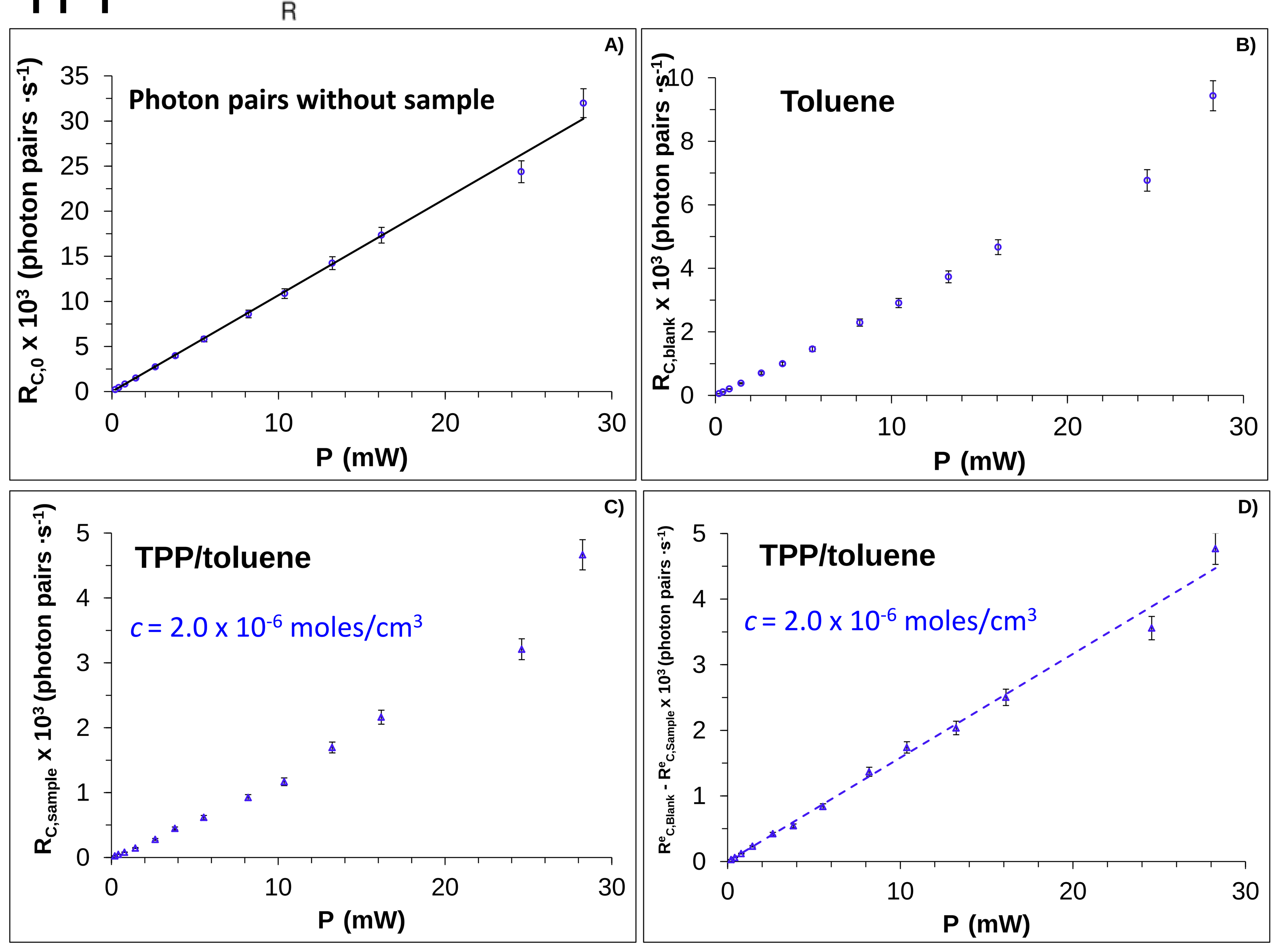
RhB



β (photons/s mW)	*c* (x 10⁻⁵ moles/cm³)	σ_E @ 810 nm (x 10⁻²¹ cm²/molecule)
59.0 ± 5.9	1.2 ± 0.06	5.2 ± 0.78
181.8 ± 18.1	12 ± 0.6	1.8 ± 0.27



TPP



β (photons/s mW)	*c* (x 10⁻⁵ moles/cm³)	σ_E @ 810 nm (x 10⁻²¹ cm²/molecule)
158.9 ± 15.6	0.2 ± 0.01	60 ± 9

Conclusions and perspectives

- An experimental setup which can be used to determine entangled two-photon absorption cross section of molecules in solution has been proposed and implemented.
- The entangled two-photon absorption process on Rhodamine-B and *meso*-Tetraphenylporphyrin in methanol and toluene respectively has been studied using entangled photons with a central wavelength around 810 nm and their respective σ_E values were estimated.
- Current experiments are focused on measuring σ_E for molecules that have well-known values of such property.
- Our setup can be used to perform ETPA measurements in a set of molecules that can be used to design new materials for entangled photon sensors[3]

References

1. Saleh, B.; Teich, M. C. *et al. Phys. Rev. Lett.* **1997,** *78*, 1679.
2. Dong-Ik, L and Goodson, T III. *J. Phys. Chem. B*. **2006,** *110*, 25582.
3. Goodson, T III.; Baurle, P. *et al. J. Am. Chem. Soc.* **2009,** *131*, 973.