

# Classical and Entangled Two-Photon Absorption in Atoms and Molecules

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## Abstract

Two-photon transitions can be excited using light with different properties such as laser light, pseudo-thermal light and entangled photons pairs (ETPA). In particular, the ETPA process has promising applications in different fields due to its linear dependence with the photon flux. Results of different ETPA experiments in organic molecules have many questions regarding the difficulty to distinguish an ETPA signal from single photon losses. In order to understand the characteristics of the TPA process with different light sources, the classical TPA cross section must be measured. In this work we present experimental values of the classical TPA cross section for cesium atoms. These are compared with theoretical ones obtained by means of second order perturbation theory. These values are the starting point for experiments of ETPA in cesium atoms. Additionally, measurements of the ETPA cross section in RhB using a coincidence detection scheme are presented. The ETPA cross section is quantified considering the effects of single photon losses in the experiment and considering parameters that are linear-loss independent. The results suggest that it is possible to witness an ETPA signal when measuring coincidences.

## Two-Photon Absorption (TPA)

### MOTIVATION

- Characterization of atoms and organic molecules.
- Starting point for studying multiphoton processes with different light sources.
- Spectroscopic information for Cesium atoms available
- Two-photon transition studied for atomic clocks and Four-Wave Mixing processes.
- Classical TPA has a quadratic dependence with photon flux.

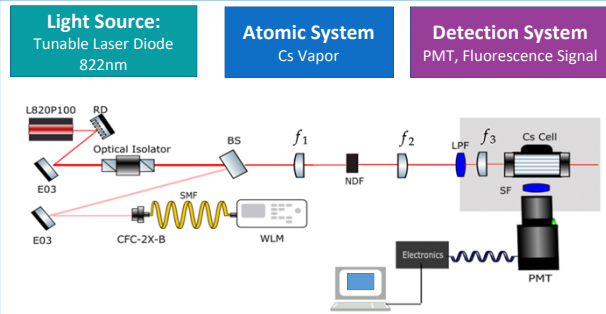
$$R_{TPA} = \delta_C \phi^2 + \sigma_E \phi$$

## Entangled Two-Photon Absorption (ETPA)

### MOTIVATION

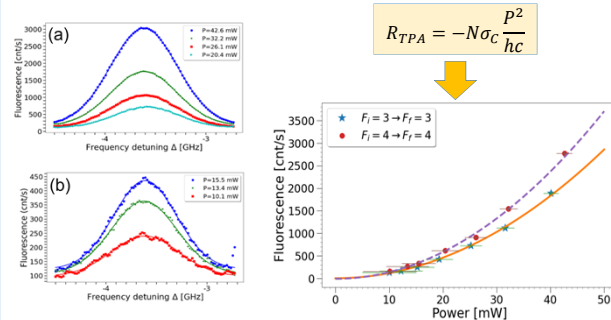
- Linear dependence with the photon flux: Enhancement of the TPA process.
- Possible application with biological samples.
- New spectroscopic techniques
- Measurements sensitive to single photon losses
- Relation between ETPA cross section and classical TPA:  $\sigma_E = \frac{\delta_C}{2A_E T_E}$

## TPA Cross-Section of Atomic Cesium



$$\sigma_C = \left( \frac{ve^4 \mu_0}{\epsilon_0 h^3} \right) \sum_m \frac{1}{3} \frac{|\langle \psi_f | \hat{\epsilon} \cdot \hat{r} | \psi_m \rangle \langle \psi_m | \hat{\epsilon} \cdot \hat{r} | \psi_i \rangle|^2}{v_m - v_i - v} \times S_{m,n} \left( \frac{\gamma_{fi}}{(v_{fi} - 2v)^2 + \gamma_{fi}^2/4} \right)$$

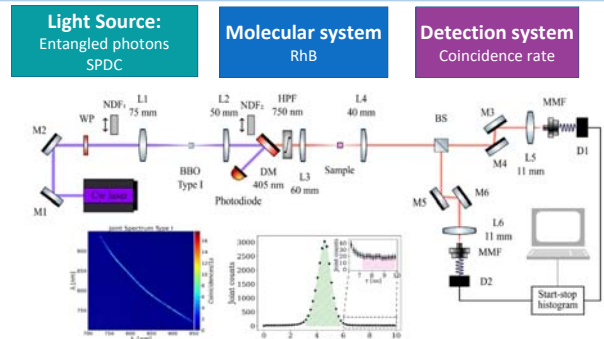
- Classical TPA cross section calculated using second order perturbation theory.
- Signal detected via Two-Photon Induced Fluorescence
- TPA excited with a CW laser at 822 nm.



Classical TPA Cross-Section	$6S_{1/2} \rightarrow 8S_{1/2}$ $F_i = 3 \rightarrow F_f = 3$	$6S_{1/2} \rightarrow 8S_{1/2}$ $F_i = 4 \rightarrow F_f = 4$
Theory Doppler-Free [ $cm^4/W$ ]	$5.3 \times 10^{-23}$	$6.3 \times 10^{-23}$
Theory Doppler broadened [ $cm^4/W$ ]	$0.99 \times 10^{-24}$	$1.2 \times 10^{-24}$
Experiment [ $cm^4/W$ ]	$(1.8 \pm 1.1) \times 10^{-24}$	$(2.2 \pm 1.1) \times 10^{-24}$

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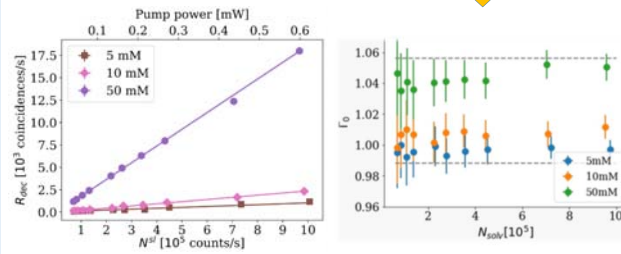
## ETPA Cross-Section of Rhodamine B



- Photon pairs from a Type I BBO crystal
- Transmission measurements with coincidences
- Sensitive to single photon losses.

$$R_{TPA} = \frac{cVN_A\sigma_E}{A} R_{solv}$$

$$\Gamma_0 = \frac{(R_1^{solv} R_2^{solv} / R_{12}^{solv})}{(R_1^{solv} R_2^{solv} / R_{12}^{solv})}$$



Concentration [mM]	ETPA Cross-Section $\sigma_E$ [ $\times 10^{-21} cm^2/molecule$ ]
5	$5.2 \pm 1.5$
10	$6.3 \pm 0.8$
50	$9.8 \pm 0.5$

A.M Torres, et al. To be Submitted.

## Conclusions and Future Work

- We calculated and measured the classical TPA cross section,  $\delta_C$ , in cesium atoms.
- We are able to witness ETPA in RhB molecules using a coincidence detection scheme.
- New experiments regarding ETPA in cesium atoms are plan for next months.

## References

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