Universidad de los Andes | Facultad de Ciencias Departamento de Física

Óptica Cuántica Uniandes

Non-Degenerate Two-Photon Absorption Cross-Section in Cesium Atoms

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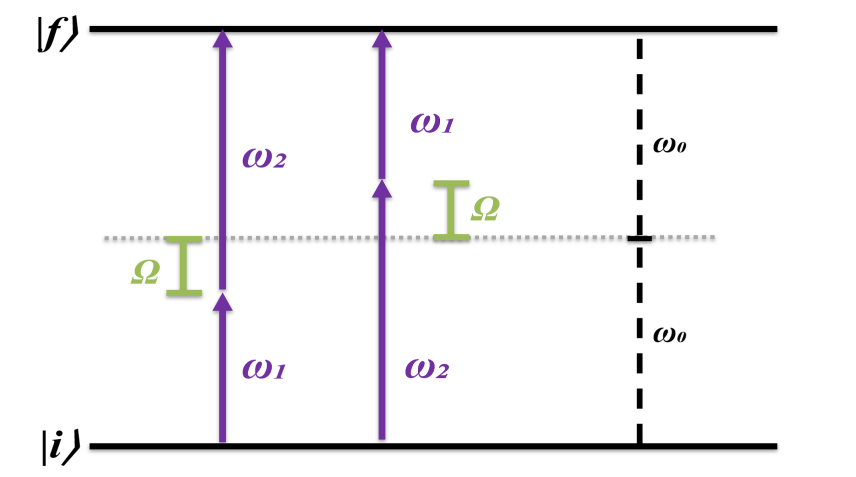
Abstract

Non-degenerate two-photon absorption (ND-TPA) offers a way to modify the absorption cross section by redistributing the excitation energy between two photons of different frequencies. We theoretically studied the 6S1/2 →8S1/2 two-photon transition in cesium by deriving the non-degenerate TPA cross section as a function of the detuning parameter Ω. The model includes the coherent contribution of the two possible absorption orderings and the intermediate nP states. Numerical results show that tuning Ω can strongly enhance the cross section when one photon approaches an intermediate resonance, while local minima arise from destructive interference between different excitation pathways. Near the resonances, the peaks follow a Lorentzian profile, with power-law behavior in the tails. At Ω=0, the model recovers the degenerate structure, although the calculated value remains lower than the reported degenerate result because hyperfine and angular factors are not yet included. These results suggest that ND-TPA could be used to identify frequency configurations that increase fluorescence counts in future cesium experiments.

Non-Degenerate Two-Photon Absorption (ND-TPA)

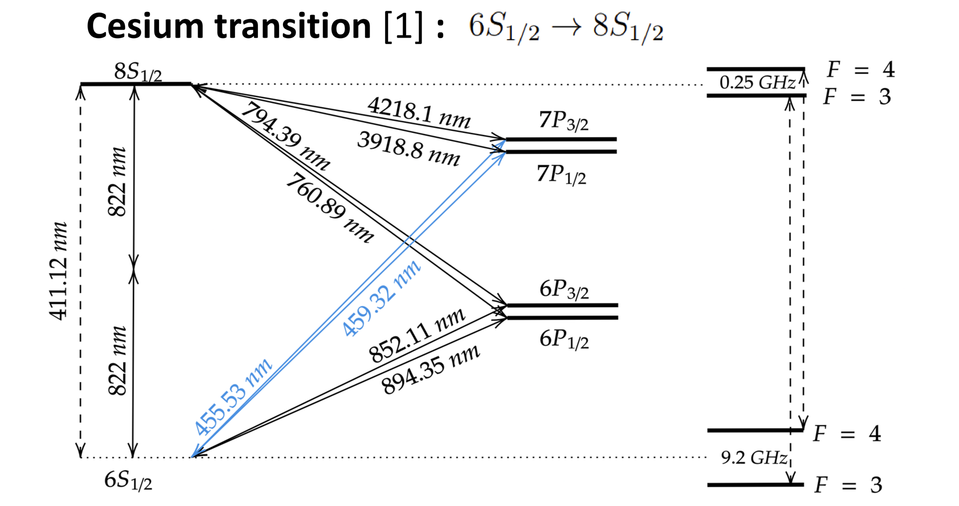
MOTIVATION

- ND TPA can **enhance the absorption cross** section by tuning the detuning parameter Ω near intermediate resonances, leading to **higher fluorescence counts** and **improved experimental detectability.**
- This work supports ongoing cesium TPA measurements in the Uniandes Quantum Optics Group [2] and provides a **theoretical basis** for moving from degenerate TPA to **non-degenerate TPA**, and ultimately to **non-degenerate ETPA**.



$\omega_1 = \omega_0 - \Omega \;;\; \omega_2 = \omega_0 + \Omega \;\}\; \omega_1 + \omega_2 = 2\omega_0 = \omega_{fi}$

Cesium transition [1] : $6S_{1/2} \rightarrow 8S_{1/2}$



Derivation of TPA Cross-Section

1. Time-Dependent Perturbation Theory

The atom-field interaction is treated in the electric dipole approximation [3]: $\hat{H}_{int}(t) = -e\hat{\mathbf{r}} \cdot \mathbf{E}(t).$

The time evolution is described TDPT, and the second-order term is the first one that contributes to TPA [5]:

$$\hat{U}^{(I)} = 1 - \frac{i}{\hbar}\int dt'\, \hat{H}^{(I)}_{int}(t') + \left(\frac{-i}{\hbar}\right)^2 \int dt' \int dt''\, \hat{H}^{(I)}_{int}(t')\hat{H}^{(I)}_{int}(t'')$$

With the transition amplitude:

$$A^{(2)}_{i\rightarrow f}(t) = \langle \psi_f |\, \hat{U}^{(I)}(t, t_0)\, |\psi_i\rangle\,.$$

2. Non-Degenerate Electric Field

In non-degenerate TPA, the field contains two frequencies:

$$\mathbf{E}(t) = \frac{E_1}{2}\left(e^{-i\omega_1 t} + e^{i\omega_1 t}\right)\hat{\epsilon}_1 + \frac{E_2}{2}\left(e^{-i\omega_2 t} + e^{i\omega_2 t}\right)\hat{\epsilon}_2.$$

For absorption, we keep the terms proportional to $e^{-i\omega_j t}$:

$$\hat{H}_{\rm int}(t) = -\frac{eE_1}{2}\hat{\epsilon}_1 \cdot \hat{\mathbf{r}}\, e^{-i\omega_1 t} - \frac{eE_2}{2}\hat{\epsilon}_2 \cdot \hat{\mathbf{r}}\, e^{-i\omega_2 t}.$$

3. Transition Amplitude

The atom can absorb one photon of each frequency in two possible time orderings: $A^{ND}_{i\rightarrow f}(t) = A_{12}(t) + A_{21}(t).$

The transition amplitude has a Two-Photon resonance with ω_{fi} and a One-Photon resonance with ω_{mi}.

4. Probability and Transition Rate

Because both paths reach the same final state, the transition probability is: $p_{ND}(t) = |A_{12} + A_{21}|^2$

$$p_{ND}(t) = |A_{12}|^2 + |A_{21}|^2 + \underbrace{2\mathrm{Re}\left[A_{12}A^*_{21}\right]}_{\text{Interference}}$$

The transition rate [1] is obtained from: $\Gamma_{ND} = \frac{dp_{ND}}{dt} \longrightarrow \sigma_{ND} = \frac{2\hbar\epsilon_0\Gamma_{ND}}{I_1 I_2}$

5. Non-Degenerate TPA Cross Section

$$\sigma_{ND}(\omega_1,\omega_2) = \frac{\mu_0 e^4 \omega_1\omega_2}{2\epsilon_0\hbar^3}\Bigg\{ \frac{\gamma_f}{(\omega_{fi}-\omega_1-\omega_2)^2+\gamma_f^2/4}\left|\sum_{m,m'}\left[\frac{\mu^{(1)}_{fm}\mu^{(2)}_{mi}}{\omega_{mi}-\omega_2} + \frac{\mu^{(2)}_{fm'}\mu^{(1)}_{m'i}}{\omega_{m'i}-\omega_1}\right]\right| + \sum_m \frac{\gamma_m}{(\omega_{fm}-\omega_1)^2+\gamma_m^2/4}\left|\sum_{m'}\frac{\mu^{(1)}_{fm'}\mu^{(2)}_{m'i}}{\omega_{m'i}-\omega_2}\right|^2$$

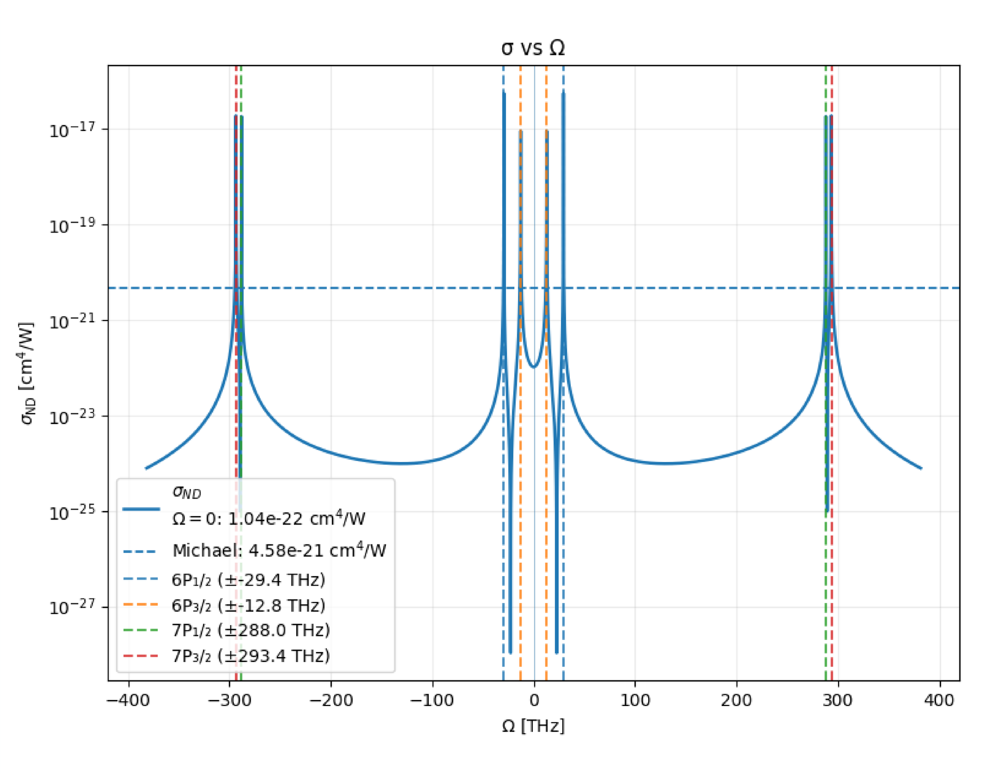
$$+\sum_m \frac{\gamma_m}{(\omega_{fm}-\omega_2)^2+\gamma_m^2/4}\left|\sum_{m'}\frac{\mu^{(2)}_{fm'}\mu^{(1)}_{m'i}}{\omega_{m'i}-\omega_1}\right|^2 + 2\,\mathrm{Re}\sum_{m,m'}\left[\frac{\left(\mu^{(1)}_{fm}\mu^{(2)}_{mi}\right)^*\mu^{(2)}_{fm'}\mu^{(1)}_{m'i}}{(\omega_{mi}-\omega_2)(\omega_{m'i}-\omega_1)}\frac{\gamma_{mm'}}{(\omega_{fm'}-\omega_1-\omega_{fm}+\omega_2)^2+\gamma^2_{mm'}/4}\right]\Bigg\}$$

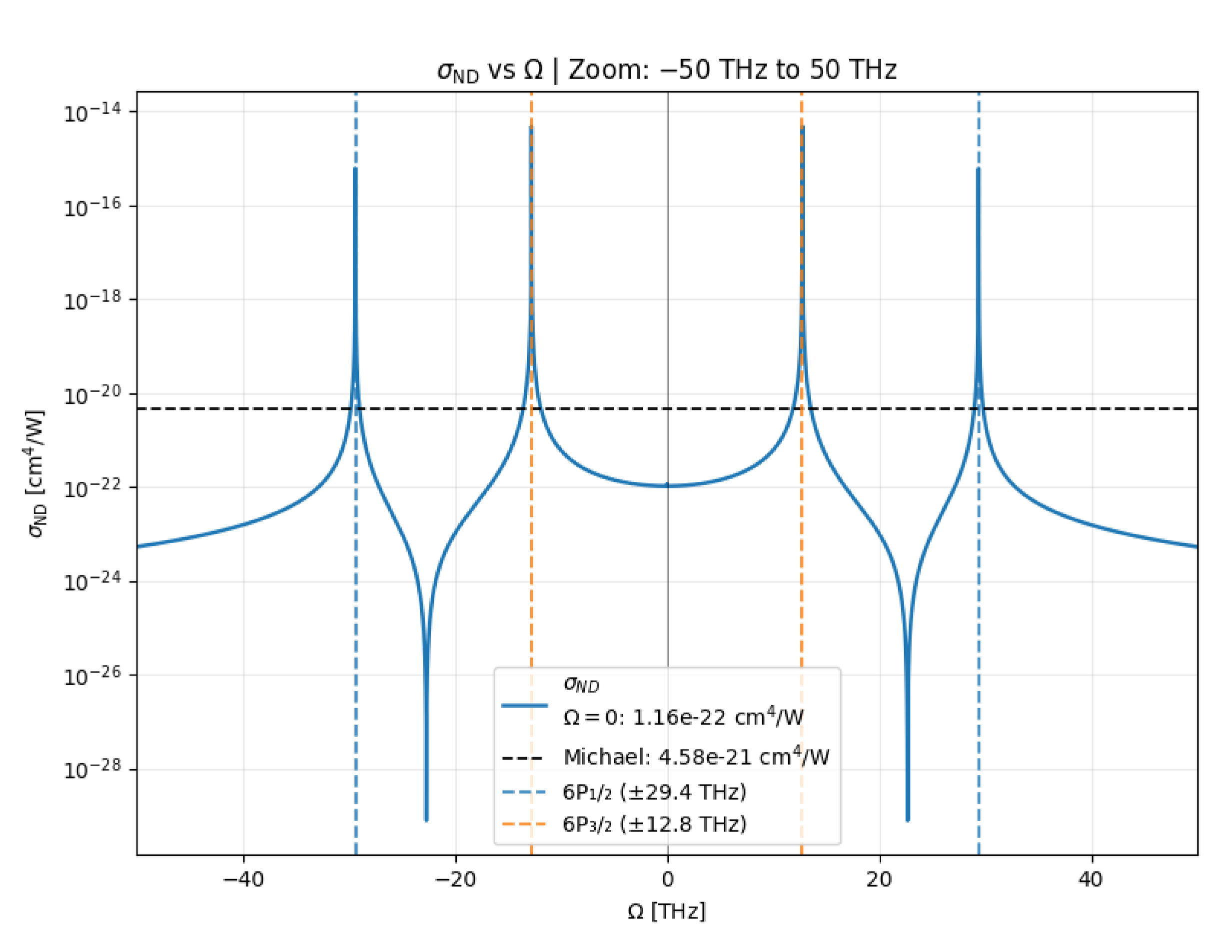
- Global two-photon resonance $\omega_1 + \omega_2 \simeq \omega_{fi}$
- Intermediate resonance via ω_1: $\omega_1 \simeq \omega_{fm}$
- Intermediate resonance via ω_2: $\omega_2 \simeq \omega_{fm}$
- Quantum-path interference

Numerical Results in Cesium 6s-8s Transition

1. Cross-Section as a function of Ω

The ND cross section was plotted as a function of Ω to identify detuning values that enhance the TPA signal.





- **Cross-section enhancement** appears when one photon approaches a real intermediate transition: $\omega_1 \simeq \omega_{mi}$ or $\omega_2 \simeq \omega_{mi}.$
- A **100 GHz detuning** from the $6P_{1/2}$ intermediate resonance gives an **enhancement of ≈3.4 orders of magnitude** relative to Ω = 0.
- **Local minima** arise from destructive interference between intermediate state paths (Fano Valleys) [7].
- Close to a peak, the dominant behavior is **Lorentzian**: $\sigma_{ND}(\delta) \propto \frac{1}{\delta^2 + \gamma_m^2/4}, \quad \delta = \omega_{fm} - \omega_j$
- At **Ω = 0**, our model obtains: $\sigma_{ND}(0) = 1.038 \times 10^{-22}\ \mathrm{cm^4/W}.$ While [1] reported the degenerate value: $\sigma_D = 4.58 \times 10^{-21}\ \mathrm{cm^4/W}.$

This difference is expected since hyperfine and angular factors are not yet included.

Conclusions and Future Work

- The model shows that **tuning Ω can enhance the Non-Degenerate TPA Cross-Section** when one photon approaches an intermediate nP resonance (**3.4 orders of magnitude** for a 100 GHz detuning).
- **Future theoretical work** should include hyperfine structure, angular factors, Wigner 6j symbols, and the effect of different polarizations or temporal orderings.
- **Experimentally**, the next step is to perform ND TPA measurements by **scanning Ω with two frequency-controlled beams** and test whether the predicted cross-section enhancement increases fluorescence counts.
- This enhancement helps identify optimal detunings for **future ETPA experiments**, where the signal is expected to be much weaker due to the low photon flux of entangled-light sources [8].

References

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