Ab-initio quantum dynamic imaging using circularly polarized light

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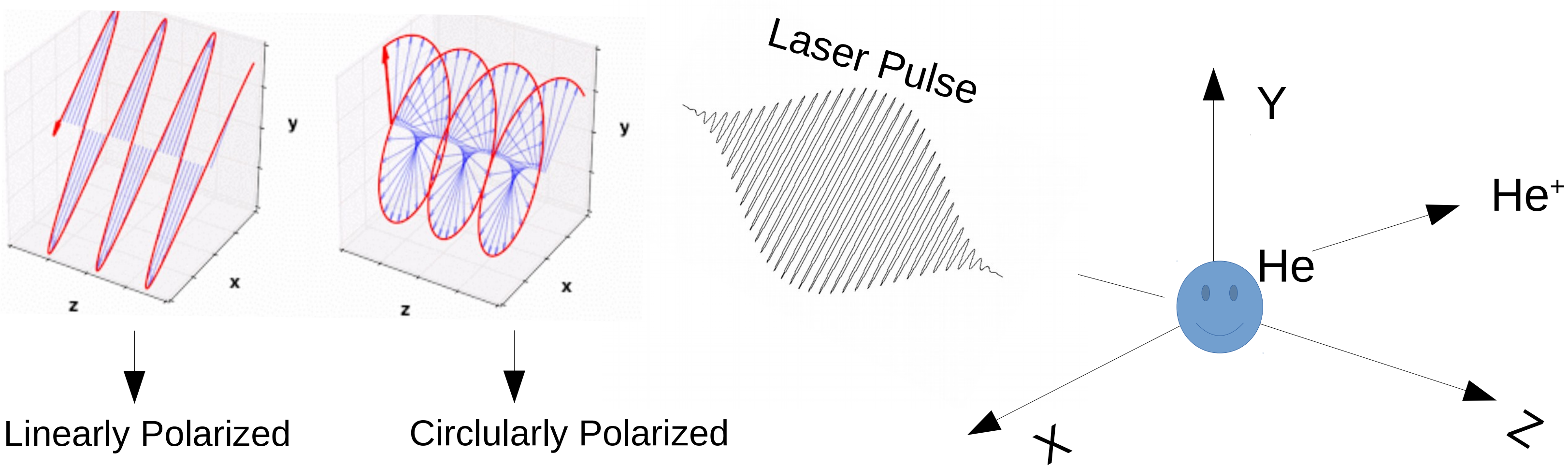
Introduction

- Finding an Ab-initio solution to the Time Dependent Schrodinger Equation (TDSE) to describe the dynamics of a multi electron atomic system.

Scenario:



- Linearly polarized light ionizes an atom without changing the magnetic quantum number of the electron i.e. $\Delta M_L = 0$
- Circularly polarized light can access continuum states with different magnetic quantum numbers as $\Delta M_L = \pm 1$ and hence more detailed structural information can be obtained.

Theoretical procedure

TDSE → $i\partial_t \psi(\vec{r}_1, \vec{r}_2; t) = [\hat{H} + \hat{D}(t)]\psi(\vec{r}_1, \vec{r}_2; t)$

$\hat{D}(t) = -A(t)[\vec{p_1} + \vec{p_2}]$

$$\psi(\vec{r}_1, \vec{r}_2; t) = \sum_{nLM_L} C_{nLM_L}(t)\phi_{nLM_L}(\vec{r}_1, \vec{r}_2)$$

Field free atomic eigen states

$$\hat{H}\phi_{nLM_L} = E_{nLM_L}\phi_{nLM_L}$$

Using *orthonormality* of the ***ϕ***'s (the two electron eigen wavefunctions):

→ $$\langle \phi_{nLM_L} | \phi_{n'L'M'_{L'}} \rangle = \delta_{nLM_L;n'L'M'_{L'}}$$

and introducing the following notation:

Represents the state of polarization

→ $$\langle \phi_{nLM_L} | \hat{D}^q | \phi_{n'L'M'_{L'}} \rangle = \hat{D}^q_{nLM_L;n'L'M'_{L'}}$$

$$i\dot{C}_{nLM_L}(t) = C_{nLM_L}(t)E_{nLM_L} + \sum_{n'L'M'_{L'}} C_{n'L'M'_{L'}}(t)\hat{D}^q_{nLM_L;n'L'M'_{L'}}$$

$$C = C^R + iC^I$$

$$\dot{C}^R = EC^I - \sum DC^R$$

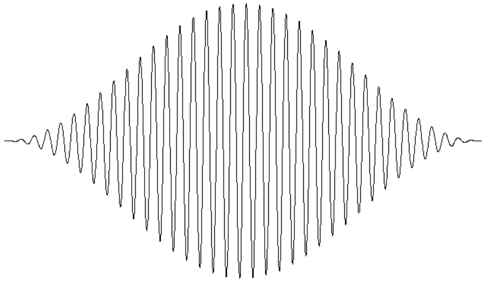
$$\dot{C}^I = -EC^R - \sum DC^I$$

$$t \geq \tau_p$$

$$IY = 1 - \sum_{nLM_L}^{E_{nLM_L} \leq E_{IP}} |C_{nLM_L}(t)|^2$$

Ionization Yield

Shape of the pulse



- Form of the laser pulse is:

$$A(t) = A_o f(t) \sin(\omega t)$$

$$f(t) = \cos^2(\pi t/\tau_p) \quad \frac{-\tau_p}{2} \le t \le \frac{\tau_p}{2}$$

$$\tau_p = n_c T_o$$

- τ_p is the total duration of the pulse which has n_c number of optical cycles.

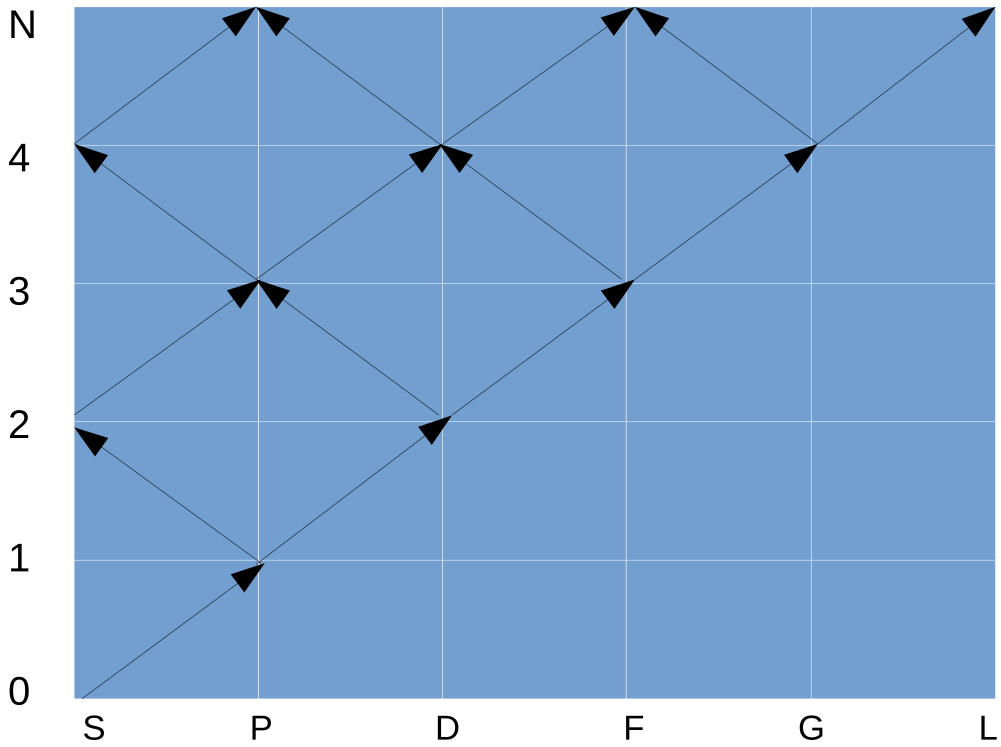
- Physical field E(t) is relatd to A(t) in dipole approximation as:

$$E(t) = -\dot{A}(t)$$

The state of polarization

- For linear polarisation: $(\mathrm{q}=0;\ \mathrm{L}'=L\pm 1; M'_{L'}=M_L)$

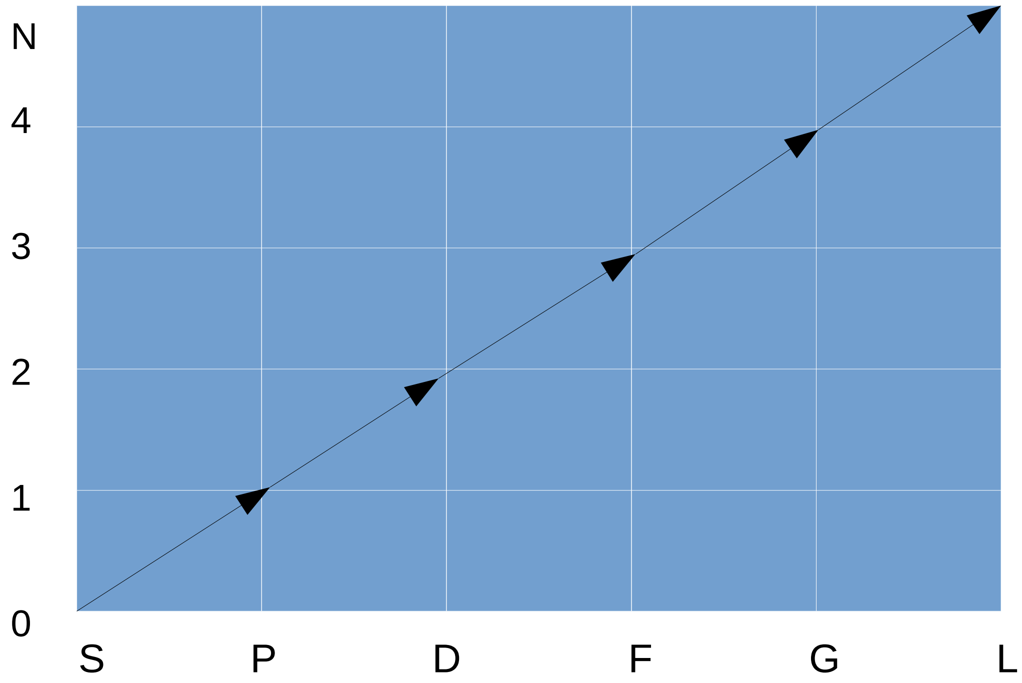
$$D^{q=0}_{nLM_L;n'L'M'_{L'}} = D_{nLM_L;n'(L\pm 1)M_L}$$



- For Circular Polarisation: $(\mathrm{q} = \pm 1; L' = L+1; M'_{L'} = M_L \mp 1)$

$$D^{q=-1}_{nLM_L;n'L'M'_{L'}} = D_{nLM_L;n'L+1M_L+1} \longrightarrow \text{RCP}$$

$$D^{q=+1}_{nLM_L;n'L'M'_{L'}} = D_{nLM_L;n'L+1M_L-1} \longrightarrow \text{LCP}$$



Thus far ...

- The physical scenario
- TDSE
- An approach to solve the TDSE
- Involvement of laser matter interaction operator D^q
- The form of the pulse used in D^q
- How the state of polarisation (q = 0, 1, -1) effects the transitions

Results

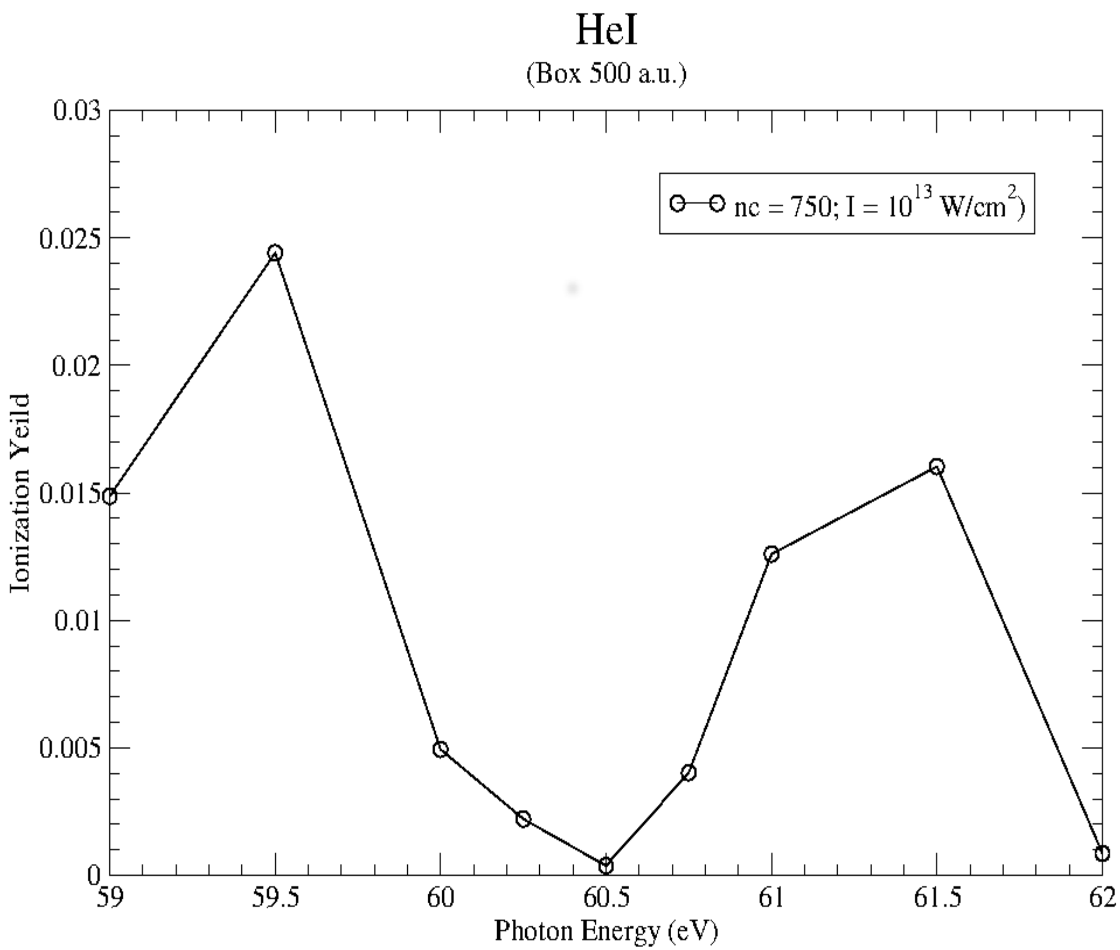
LP:

- Used linearly polarized laser beam to check the ionization behavior.
- Compared the results of TDSE with that of widely used perturbation theory.

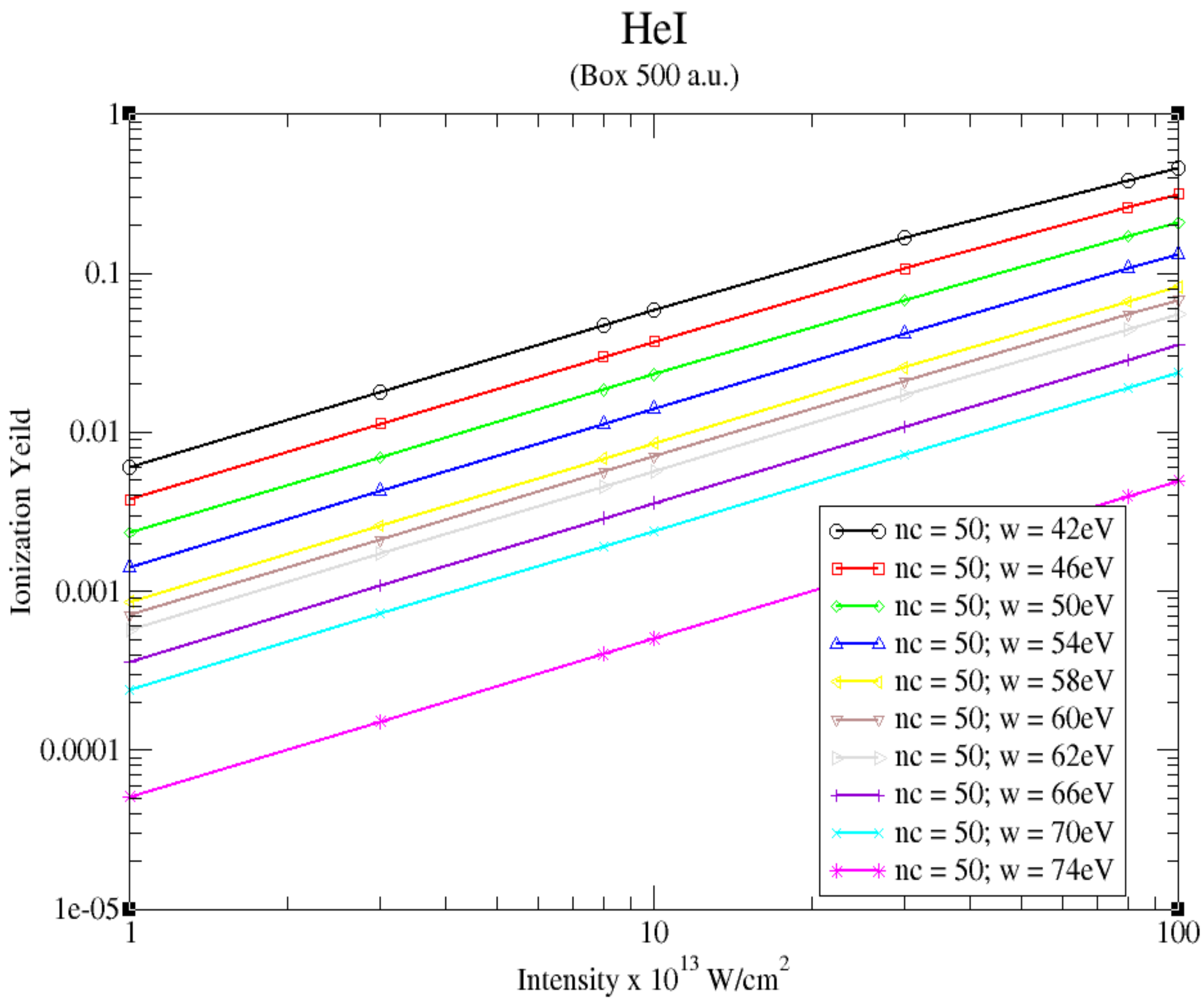
LP Vs CP:

- Incorporated circular polrization.
- Compared the ionization yields from circularly polarized and linearly polarized laser beams.

Ionization Yield Vs Photon Energy



Ionization Yield Vs Intensity



- Comparing the one photon crossection results obtained from solving the TDSE with that of perturbation theory.

$$\sigma_1 = IY_1(\frac{\omega}{I})\frac{1}{\tau_1} \qquad \tau_1 = \frac{3}{8}\tau_p$$

ω(eV) (10^{13}W/cm^2)	IY (TDSE)	σ_1 (TDSE)	σ_1 (Perturbation)	Δ(%)
42	6.04453	2.203	2.186	0.64
46	3.76076	1.644	1.636	0.49
50	2.36709	1.222	1.208	1.14
54	1.43904	0.866	0.834	3.6

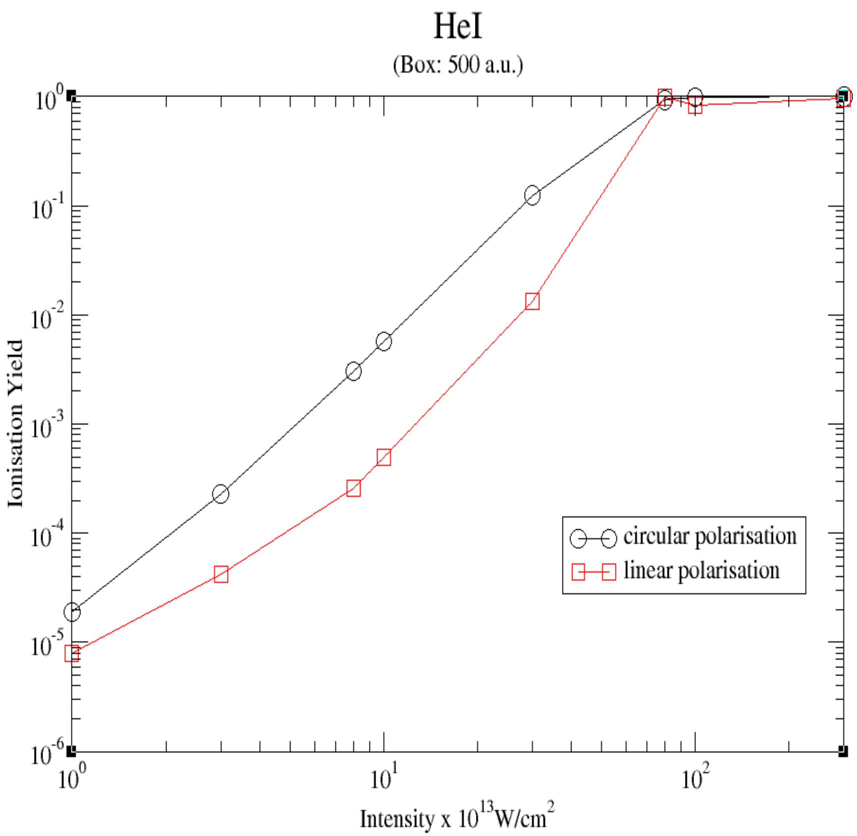
- Comparing the three photon crossection results obtained from solving the TDSE with that of perturbation theory. Photon energy used is 10eV.

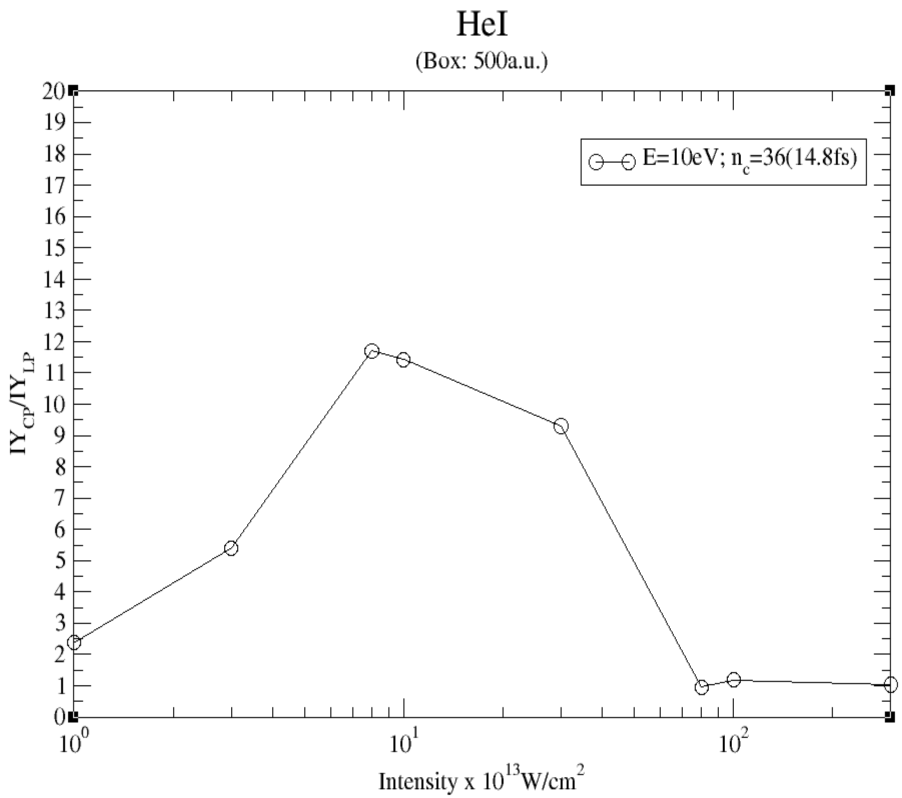
$$\sigma_3 = IY_3(\frac{\omega}{I})^3 \frac{1}{\tau_3} \qquad \tau_3 = \frac{231}{1024}\tau_p$$

Intensity (x 10^{13}W/cm^2)	IY_3 (TDSE)	σ_3(TDSE) x 10^{-85} cm^{6}s^2	σ_3 (Perturbation) x 10^{-85} cm^{6}s^2	Δ(%)
1	0.806	98	6.5	93.36
3	4.2276	19.2	6.5	66
8	25.858	6.195	6.5	4.92
10	50.052	6.14	6.5	5.86
30	1341.9	6.097	6.5	6.6
80	97654	23.39	6.5	72.21
100	83640	10.26	6.5	36.65

LP Vs CP

- Ionization yield comparision between LP and CP laser beams.
- Photon energy is 10eV for both the polarizations (three photon ionization)





Future work

- Next step is to include fluctuations in the electric field (FEL radiation).
- Yileds and Angular Distrubutions will be calculated.

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Thank You