# One, Two and Three Photon Absorption in Atomic Hydrogen

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#### **Abstract**

Non relativistic, semi-classical in dipole approximation, based on time dependent Schrodinger equation under the framework of perturbation theory, three different absorption processes in hydrogen atom are studied and calculated transition matrix element from an initial state to final state.

**Key words:** multiphoton ionization, perturbation and ionization rate

#### Introduction

Multiphoton ionization results from the simultaneous absorption of several photons, predicted(Koval 2004) theoretically in 1931 by M. Geoppert, were for a long time considered to be mainly of academic interest. This view changed when it was shown (Delone, Mainfray, Charles) that a two-photon absorption process could, because of a quadratic dependence of excitation on intensity, produce a spatially confined excitation useful for three-dimensional data storage and imaging. Two-photon absorption has received considerable attention recently because of the development of highly efficient two-photon-sensitive materials, leading to numerous technological applications. These successes have created interest in exploring applications based on three-photon excitations (3PA)(Joseph 2008). 3PA works when three photons (identical or different frequencies) are simultaneously absorbed in one event and make a transition from initial state |g| to and excited state or final state  $|f\rangle$  allowed by three photon selection rules (Thayyullathil et al. 2003). The energy difference between the involved lower and upper states of the atom is equal to the sum of the energies of the three photons. The three photon absorption most commonly occurs in longer wavelengths (near infrared), some scientists see hope for it in terms of biomedical and photonic applications (Joseph 2008).

Multiphoton absorptions are described by higher order perturbation theory, which is valid when the radiation field strength is less than the atomic field strength. If a quantum system is represented by the time dependent Hamiltonian H, then from TDSE (Peter et al. 2005).

$$H|\Psi(t)\rangle = E|\Psi(t)\rangle$$
 (1)

Here H is the total Hamiltonian, which is the sum of unperturbed Hamiltonian  $H_0$  and interaction Hamiltonian H'(t) such as;

 $H = H_0 + H'(t)$ H'(t) has the form

$$H'(t) = H'(t)$$
 if  $0 \le t \le \tau$   
= 0 if,  $t \le 0$  and,  $t \ge \tau$  (2)

Assuming the solution of this equation to be linear combination of basis set  $\{|n\rangle\}$ , which are the engienstates of the unperturbed Hamiltonian,

$$|\Psi(t)\rangle = \sum_{n} C_{n}(t)\varphi_{n}(r)e^{-iE_{n}t/\hbar}$$
 (3)

Unperturbed Hamiltonian Ho has eigenvalues as:

$$H_0|n\rangle = E_n|n\rangle$$
 (4)

The quantities to be determined here are the expansion coefficient  $C_n(t)$  and these are our direct physical interest.  $\varphi_n(r)$  is the eigenfunction of the Hamiltonian. The square of this expansion coefficient is the probability, and divided by time is the transition rate ( $\Gamma_{fa}$ ).

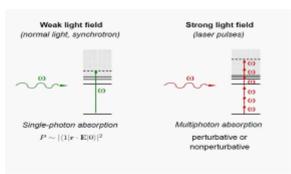


Fig. 1. Laser - Matter Interaction

 $|C_n(t)|^2$ , with which is the system is described by  $|\Psi(t)\rangle$  will be found in the energy eigenstate  $|n\rangle$  at time t. Using atomic units as  $\hbar=m=e=a_0=1$ ; We can write, interaction Hamiltonian for three different laser frequency such as:

$$H' = \sum_{i=1to3} \hat{\varepsilon}_i \cdot \vec{r} E_{0i} e^{i(\omega_j t - \delta_j)}$$
 (5)

$$H' = \vec{\epsilon_1} \cdot \vec{\tau} \vec{E}_{01} e^{-i(\omega_1 t + \delta_2)} + \vec{\epsilon_2} \cdot \vec{\tau} \vec{E}_{02} e^{-i(\omega_2 t + \delta_2)} + \vec{\epsilon_3} \cdot \vec{\tau} \vec{E}_{02} e^{-i(\omega_2 t + \delta_2)} (6)$$

Let,  $H_1 = \hat{\varepsilon}_1 \cdot \vec{r} E_{01}$  Similarly we can write for  $H_2$  and,  $H_3$ . Here,  $E_{01}$  is the amplitude of the electromagnetic field along the direction of the polarization vector  $\hat{\varepsilon}_1$ . Similarly  $E_{02}$  and  $E_{03}$  are the amplitudes along the direction  $\hat{\varepsilon}_2$  and  $\hat{\varepsilon}_3$  for the beam 2 and 3 respectively. The interaction Hamiltonian for the same laser frequency  $\omega$  with different polarization should be:

$$H' = \sum_{j} \hat{\varepsilon}_{j} \cdot \vec{r} E_{0} e^{i(\omega t + \delta_{j})}$$
 (7)

#### One-photon ionization

The transition of electron from ground state to the final state by the absorption of one photon Singlephoton absorption is described by first order perturbation theory.

#### Calculation for first order perturbation:

$$\frac{dC_n^{(1)}(t)}{dt} = \frac{1}{i} \sum_g C_g^{(0)}(t) e^{i\omega_{ng}t} \langle n|H'|g \rangle e^{-i\omega t}$$
 (8)

Here,  $\omega$  is the frequency of the linearly polarized light. The ionization rate for the same frequency and different polarization can be expressed as (Shrestha et al. 2008):

$$\frac{\left|c_{n}^{(1)}(t)\right|^{2}}{t}=E_{0}^{2}\left|\langle n|\hat{\varepsilon_{j}}\cdot\vec{r}|g\rangle\right|^{2}\delta(\omega_{ng}-\omega)$$

Since atoms are small compared to the wavelength of light, the amplitudes of electric field will not vary significantly over the dimensions of atom. We can therefore take them constants out from the calculation of integrals (Mark 2006). Thus,

$$\Gamma_{ng}^{(1)} \otimes |\langle n|H'|g\rangle|^2 \delta(\omega_{ng} - \omega)$$

Here we can see that a resonance occurs at  $\omega_{ng} = \omega$  corresponding to one photon transition from ground state  $|g\rangle$  to the final state  $|n\rangle$ .

#### Two photon absorption (2PA)

2PA works when two photons (identical or different frequencies) are simultaneously absorbed in one event and make a transition from initial state as ground state  $|g\rangle$  to and final state  $|m\rangle$  allowed by two photon selection rules (Bethe *et al.* 1957). The two photon absorption is described by the second order perturbation theory.

$$\frac{dC_n^{(2)}(t)}{dt} = \frac{1}{i} \sum_n C_n^{(1)}(t) \langle m|H'|n \rangle e^{i(\omega_{mn} - \omega)t} \qquad (9)$$

By integrating and substituting the value of  $C_n^{(1)}(t)$  (Shrestha *et al.* 2009),

$$C_{m}^{(2)}(t) = \sum_{n} \frac{(m|H'|n)(n|H'|g)[\frac{(-a^{t}|a_{mn}-a)(a_{mn}-a)t}{(a_{mn}-a)(a_{mn}-a)}]}{\times [\frac{(-a^{t}|a_{mn}+a_{mn}-a)t}{(a_{mn}-a)(a_{mn}-a)}]}$$
(10)

Assuming the two different beams are j and k. Let  $M_{mg}^{(2)}$ , is the transition matrix element for two-photon ionization, for same frequency  $(\omega)$  of different polarization. We can derive by dropping the antiresonance terms such as

$$M_{ng}^{(2)} = \sum_{j,k} \sum_{n} \frac{\langle m | \bar{\varepsilon}_k \cdot \bar{r} | n \rangle \langle n | \bar{\varepsilon}_j \cdot \bar{r} | g \rangle}{(\omega_{ng} - \omega)}$$
(11)

The transition rate (Thayyullathil et al. 1994)

$$\Gamma_{mg}^{(2)} \propto \left| M_{mg}^{(2)} \right|^2 \delta(\omega_{mg} - 2\omega)$$

Where we have retained only those terms in which the denominators approaches zero. Here we can see that a resonance occurs at  $\omega_{ma} = 2\omega$  corresponding to the two photon transition for the identical laser frequency  $\omega$ , from the ground state  $|g\rangle$  to the final state  $|m\rangle$  through one intermediate state  $|n\rangle$ .

#### For different laser frequency with different polarization

The second order expansion coefficient for different laser frequency with different polarization:

$$C_m^{(2)}(t) = \sum_n \frac{\langle m|H_1|n\rangle\langle n|H_1|g\rangle}{\omega_{n,\sigma}-\omega_1} \delta(\omega_{mg} - 2\omega_1)$$
 (12)

$$+\sum_{n}\frac{\langle m|H_{2}|n\rangle\langle n|H_{1}|g\rangle}{(\omega_{ng}-\omega_{1})}\delta(\omega_{mn}+\omega_{ng}-\omega_{1}-\omega_{2})$$

$$+\sum_{n}\frac{\langle m|H_{3}|n\rangle\langle n|H_{1}|g\rangle}{(\omega_{ng}-\omega_{1})}\delta(\omega_{mn}+\omega_{ng}-\omega_{1}-\omega_{3})$$

$$+ \sum\limits_{n} \frac{\langle m|H_{2}|n\rangle\langle n|H_{2}|g\rangle}{(\omega_{ng}-\omega_{2})} \delta(\omega_{mn}+\omega_{ng}-2\omega_{2})$$

$$+ \sum\limits_{n} \frac{\langle m| {\it H}_{1}|n\rangle \langle n| {\it H}_{2}|g\rangle}{(\omega_{ng}-\omega_{2})} \delta(\omega_{mn}+\omega_{ng}-\omega_{1}-\omega_{2})$$

$$+\sum_{n}\frac{\langle m|H_{3}|n\rangle\langle n|H_{2}|g\rangle}{(\omega_{ng}-\omega_{2})}\delta(\omega_{mn}+\omega_{ng}-\omega_{2}-\omega_{3})$$

$$+ \sum_{n} \frac{\langle m|\mathcal{H}_{3}|n\rangle\langle n|\mathcal{H}_{3}|g\rangle}{(\omega_{ng}-\omega_{3})} \delta(\omega_{mn}+\omega_{ng}-2\omega_{3})$$

$$+ \sum_{n} \frac{\langle m|H_{1}|n\rangle\langle n|H_{3}|g\rangle}{(\omega_{ng} - \omega_{3})} \delta(\omega_{mn} + \omega_{ng} - \omega_{1} - \omega_{3}) \quad (13)$$

$$+ \textstyle\sum_{\mathbf{n}} \frac{\langle m|H_2|n\rangle\langle n|H_3|g\rangle}{(\omega_{\mathbf{n}g} - \omega_3)} \delta(\omega_{mn} + \omega_{ng} - \omega_2 - \omega_3)$$

Hence the transition matrix becomes:
$$M_{mg}^{(2)} = \sum_{j,k} \sum_{n} \frac{(m|H_k|n)(n|H_j|g)}{(\omega_{ng} - \omega_j)}$$
Three photon absorption(3PA)

3PA works when three photons (identical or different frequencies) are simultaneously absorbed in one event and make a transition from  $|g\rangle$  to excited state  $|f\rangle$ 

allowed by three photon selection rules. We require third order perturbation theory such as, for the same frequency and linearly polarized:

$$\frac{dc_f^{(3)}(t)}{dt} = \frac{1}{i} \sum_m \langle f | H' | m \rangle C_m^{(2)}(t) e^{i(\omega f_m - \omega)t}$$
(15)

Then, integrating and substituting the value of  $C_m^{(2)}$ ,

$$C_f^{(3)}(t) = \sum_m \sum_n \langle f|H'|m\rangle\langle m|H'|n\rangle\langle n|H'|g\rangle \times$$

$$\begin{bmatrix} 1-\mathrm{g}^{\mathrm{i}(\omega_{fm}-\omega)t} \\ (\omega_{fm}-\omega)(\omega_{mn}-\omega)(\omega_{mg}-\omega) \end{bmatrix} = \begin{bmatrix} 1-\mathrm{g}^{\mathrm{i}(\omega_{fm}+\omega_{mn}-2\omega)t} \\ (\omega_{fm}+\omega_{mn}-2\omega)(\omega_{mn}-\omega_{d})(\omega_{ng}-\omega) \end{bmatrix}$$

$$\left[\frac{1-e^{\int(\omega_{fm}-\omega)t}}{(\omega_{fm}-\omega)(\omega_{mg}-2\omega)(\omega_{ng}-\omega)}\right] = \frac{1-e^{\int(\omega_{fm}+\omega_{mn}+\omega_{ng}-2\omega)t}}{(\omega_{fm}+\omega_{mg}+\omega_{ng}-2\omega)(\omega_{mg}-2\omega)(\omega_{ng}-\omega)}$$

Here we can see that a resonance occurs at  $\omega_{fm} + \omega_{mn} + \omega_{ng} = \omega_{fg} = 3\omega_1$  corresponding to three photon transition for identical laser frequency  $\omega_1$ , from ground state  $|g\rangle$  to the final state  $|f\rangle$  through two intermediate states  $|n\rangle$  and  $|m\rangle$ . By dropping the antiresonance term the matrix elements for threephoton of same frequency  $(\omega)$  with different polarization becomes:

$$M_{fg}^{(3)} = \sum_{j,k,l} \sum_{m,n} \frac{\langle f(\bar{\varepsilon}_l \cdot \bar{r}) | m \rangle \langle m | (\bar{\varepsilon}_k \cdot \bar{r}) | n \rangle \langle n | (\bar{\varepsilon}_j \cdot \bar{r}) | g \rangle}{(\omega_{ng} - \omega)(\omega_{ng} - 2\omega)}$$
(17)

Similarly the matrix element for three different frequencies and polarization we have obtained 27 terms with different dirac function such as:

$$M_{fq}^{(2)} = \sum_{m} \sum_{n} \frac{\langle f | H_{4} | m \rangle \langle m | H_{4} | n \rangle \langle n | H_{4} | g \rangle}{(\omega_{mg} - 2\omega_{\perp})(\omega_{ng} - \omega_{\perp})} \delta(\omega_{fg} - 3\omega_{1})$$
 (18)

$$+\sum_{m}\,\sum_{n}\,\frac{\langle f|\mathcal{H}_{2}|m\rangle\langle m|\mathcal{H}_{1}|n\rangle\langle n|\mathcal{H}_{1}|g\rangle}{(\omega_{mg}-2\omega_{1})(\omega_{ng}-\omega_{1})}\delta\left(\omega_{fg}\,-2\omega_{1}-\omega_{2}\right)$$

$$+\sum_{m}\,\sum_{n}\,\frac{\langle f|\mathcal{H}_{3}|m\rangle\langle m|\mathcal{H}_{1}|n\rangle\langle n|\mathcal{H}_{1}|g\rangle}{(\omega_{mg}-2\omega_{1})(\omega_{ng}-\omega_{1})}\delta(\omega_{fg}\,-2\omega_{1}-\omega_{3})$$

$$+\sum_{m}\,\sum_{n}\,\frac{\langle f|\mathcal{H}_{1}|m\rangle\langle m|\mathcal{H}_{2}|n\rangle\langle n|\mathcal{H}_{1}|g\rangle}{(\omega_{mg}-\omega_{1}-\omega_{2})(\omega_{ng}-\omega_{1})}\delta\left(\omega_{fg}\,-2\omega_{1}-\omega_{2}\right)$$

$$+ \sum_{m} \; \sum_{n} \frac{(f|\mathcal{H}_{3}|m)(m|\mathcal{H}_{2}|n)(n|\mathcal{H}_{1}|g)}{(\omega_{mg}-\omega_{1}-\omega_{2})(\omega_{ng}-\omega_{1})} \delta(\omega_{fg} \; - \; \omega_{1} - \; \omega_{2} - \; \omega_{1} - \; \omega_{2} - \; \omega_{3} - \; \omega_{3} - \; \omega_{4} - \; \omega_{5} - \; \omega$$

$$+ \sum_{m} \sum_{n} \frac{\langle f|H_{2}|m\rangle\langle m|H_{2}|n\rangle\langle n|H_{1}|g\rangle}{(\omega_{mg}-\omega_{1}-\omega_{2})(\omega_{ng}-\omega_{1})} \delta(\omega_{fg}-2\omega_{2}-\omega_{1})$$

$$+\sum_{m}\sum_{n}\frac{\langle f|H_{1}|m\rangle\langle m|H_{3}|n\rangle\langle n|H_{2}|g\rangle}{(\omega_{m,n}-\omega_{2}-\omega_{2})(\omega_{m,n}-\omega_{2})}\delta(\omega_{fg}-\omega_{1}-\omega_{2}-\omega$$

$$+\sum_{m}\sum_{n}\frac{\langle f|H_{3}|m\rangle\langle m|H_{3}|n\rangle\langle n|H_{3}|g\rangle}{(\omega_{m\sigma}-2\omega_{3})(\omega_{n\sigma}-\omega_{3})}\delta(\omega_{fg}-3\omega_{3})$$

$$+ \sum_{m} \sum_{n} \frac{\langle f|\mathcal{H}_{1}|m\rangle\langle m|\mathcal{H}_{1}|n\rangle\langle n|\mathcal{H}_{3}|g\rangle}{(\omega_{mg}-\omega_{1}-\omega_{3})(\omega_{ng}-\omega_{2})} \delta(\omega_{fg}-2\omega_{1}-\omega_{3})$$

$$+\sum_{m}\sum_{n}\frac{\langle f|H_{2}|m\rangle\langle m|H_{1}|n\rangle\langle n|H_{3}|g\rangle}{(\omega_{mg}-\omega_{1}-\omega_{3})(\omega_{ng}-\omega_{3})}\delta(\omega_{fg}-\omega_{1}-\omega_{2}-\omega_{2}-\omega_{3})$$

$$+\sum_{m}\sum_{n}\frac{(f|H_{1}|m)(m|H_{3}|n)(n|H_{1}|g)}{(\omega_{mg}-\omega_{2}-\omega_{3})(\omega_{ng}-\omega_{2})}\delta(\omega_{fg}-\omega_{1}-\omega_{2}-\omega$$

$$+ \sum_{m} \sum_{n} \frac{\langle f|H_{3}|m\rangle\langle m|H_{1}|n\rangle\langle n|H_{3}|g\rangle}{(\omega_{mg}-\omega_{1}-\omega_{3})(\omega_{ng}-\omega_{3})} \delta(\omega_{fg}-\omega_{1}-2\omega_{2})$$

$$+\sum_{m}\sum_{n}\frac{\langle f|H_{1}|m\rangle\langle m|H_{3}|n\rangle\langle n|H_{1}|g\rangle}{(\omega_{mg}-\omega_{1}-\omega_{3})(\omega_{ng}-\omega_{1})}\delta\left(\omega_{fg}-2\omega_{1}-\omega_{3}\right)$$

$$+\sum_{m}\sum_{n}\frac{\langle f|H_{3}|m\rangle\langle m|H_{3}|n\rangle\langle n|H_{1}|g\rangle}{\langle\omega_{mg}-\omega_{1}-\omega_{3}\rangle\langle\omega_{ng}-\omega_{1}\rangle}\delta(\omega_{fg}-2\omega_{3}-\omega_{1})$$

$$+\sum_{m}\sum_{n}\frac{(f|H_{3}|m)\langle m|H_{1}|n\rangle\langle n|H_{2}|g\rangle}{(\omega_{mg}-\omega_{1}-\omega_{2})(\omega_{ng}-\omega_{2})}\delta(\omega_{fg}-\omega_{1}-\omega_{2}-\omega_{2})$$

$$+\sum_{m}\sum_{n}\frac{\langle f|H_{1}|m\rangle\langle m|H_{3}|n\rangle\langle n|H_{2}|g\rangle}{(\omega_{mg}-\omega_{2}-\omega_{3})(\omega_{ng}-\omega_{2})}\delta(\omega_{fg}-\omega_{1}-\omega_{2}-\omega_{3})$$

$$+ \sum_{m} \sum_{n} \frac{\langle f|H_{3}|m\rangle\langle m|H_{3}|n\rangle\langle n|H_{2}|g\rangle}{\langle \omega_{m,\sigma}-\omega_{2}-\omega_{3}\rangle\langle \omega_{n,\sigma}-\omega_{2}\rangle} \delta(\omega_{fg}-\omega_{2}-2\,\omega_{3})$$

$$+\sum_{m}\sum_{n}\frac{\langle f|H_{1}|m\rangle\langle m|H_{3}|n\rangle\langle n|H_{3}|g\rangle}{(\omega_{mg}-2\omega_{3})(\omega_{ni}-\omega_{3})}\delta(\omega_{fg}-\omega_{1}-2\omega_{3})$$

$$+ \sum_{m} \sum_{n} \frac{\langle f | \mathcal{H}_{2} | m \rangle \langle m | \mathcal{H}_{3} | n \rangle \langle n | \mathcal{H}_{3} | g \rangle}{(\omega_{m\sigma} - 2\omega_{3})(\omega_{n\sigma} - \omega_{3})} \delta(\omega_{fg} - \omega_{2} - 2\omega_{3})$$

$$+ \sum_{m} \sum_{n} \frac{\langle f|H_{1}|m\rangle\langle m|H_{2}|n\rangle\langle n|H_{3}|g\rangle}{(\omega_{mg}-\omega_{2}-\omega_{3})(\omega_{ng}-\omega_{2})} \delta(\omega_{fg}-\omega_{1}-\omega_{2}-\omega_{3})$$

$$+\sum_{m}\sum_{n}\frac{(f|\mathcal{H}_{2}|m)(m|\mathcal{H}_{2}|n)(n|\mathcal{H}_{3}|g)}{(\omega_{m\sigma}-\omega_{2}-\omega_{3})(\omega_{n\sigma}-\omega_{3})}\delta(\omega_{fg}-2\omega_{2}-\omega_{3})$$

$$\begin{split} &+ \sum_{m} \sum_{n} \frac{(f|\mathcal{H}_{3}|m)(m|\mathcal{H}_{2}|n)(n|\mathcal{H}_{3}|g)}{(\omega_{ng} - \omega_{2} - \omega_{3})(\omega_{ng} - \omega_{3})} \delta(\omega_{fg} - \omega_{2} - 2\omega_{3}) \\ &+ \sum_{m} \sum_{n} \frac{(f|\mathcal{H}_{2}|m)(m|\mathcal{H}_{3}|n)(n|\mathcal{H}_{1}|g)}{(\omega_{ng} - \omega_{1} - \omega_{3})(\omega_{ng} - \omega_{1})} \delta(\omega_{fg} - \omega_{1} - \omega_{2} - \omega_{3}) \end{split}$$

$$+\sum_{m}\sum_{n}\frac{\langle f|H_{2}|m\rangle\langle m|H_{3}|n\rangle\langle n|H_{1}|g\rangle}{\langle \omega_{n}-\omega_{n}-\omega_{n}\rangle\langle \omega_{n}-\omega_{n}\rangle}\delta(\omega_{fg}-\omega_{1}-\omega_{2}-\omega_{2}-\omega_{2}-\omega_{3})$$

$$+\sum_{m}\sum_{n}\frac{\langle f|\mathcal{H}_{2}|m\rangle\langle m|\mathcal{H}_{2}|n\rangle\langle n|\mathcal{H}_{2}|g\rangle}{(\omega_{mg}-2\omega_{2})(\omega_{ng}-\omega_{2})}\delta(\omega_{fg}-3\omega_{2}$$

$$+ \sum_{m} \sum_{n} \frac{\langle f|H_{3}|m\rangle\langle m|H_{2}|n\rangle\langle n|H_{2}|g\rangle}{(\omega_{m\sigma} - 2\omega_{2})(\omega_{n\sigma} - \omega_{2})} \delta(\omega_{fg} - 2\omega_{2} - \omega_{3})$$

$$+\sum_{m}\sum_{n}\frac{\langle f|H_{1}|m\rangle\langle m|H_{1}|n\rangle\langle n|H_{2}|g\rangle}{(\omega_{mg}-\omega_{1}-\omega_{2})(\omega_{ng}-\omega_{2})}\delta(\omega_{fg}-2\omega_{1}-\omega_{2})$$

$$+\sum_{m}\sum_{n}\frac{\langle f|H_{2}|m\rangle\langle m|H_{1}|n\rangle\langle n|H_{2}|g\rangle}{(\omega_{mg}-\omega_{1}-\omega_{2})(\omega_{ng}-\omega_{2})}\delta(\omega_{fg}-2\omega_{2}-\omega_{1})$$

We can expressed representing three different beams j, k, and l as:

$$M_{fg}^{(3)} = \sum_{i,k,l} \sum_{m,n} \frac{\langle f | \hat{\epsilon}_l \cdot \vec{r} | m \rangle \langle m | \hat{\epsilon}_k \cdot \vec{r} | n \rangle \langle n | \hat{\epsilon}_j \cdot \vec{r} | g \rangle}{(\omega_{n\sigma} - \omega_i)(\omega_{m\sigma} - \omega_i - \omega_k)}$$
 (19)

Where we have retained only those terms in which the denominators approaches zero. Here we can see resonance  $\omega_{fm} + \omega_{mn} + \omega_{ng} = \omega_{fg} = 3\omega$  corresponding three photon transition from the ground state  $|g\rangle$  to the final state  $|f\rangle$  through two intermediate states  $|n\rangle$  and  $|m\rangle$  processes. Therefore the rate of three photon absorption becomes

$$\Gamma_{fg}^{(3)} \infty |M_{fg}^{(3)}|^2 \delta(\omega_{fg} - 3\omega)$$
 (20)

#### **Results and Discussion**

- In general, all the transitions which satisfies the Bohr's quantum condition (for the single photon) are not allowed.
- The radiative transitions can take place only the states allowed by certain selection rules. The parity rule follows by odd parity of dipole operator.
- The spin selection rules follows from the fact that, the photon does not interact with electron spin and so the spin quantum number never change in the transitions.
- In a non linear medium two and three photon absorptions occurs between the states of same parity, while in the single photon absorption states involved are opposite parity. Hence as the intensity of the radiation field increases, the transition which is forbidden by single photon transitions can occur by two and three photon absorption.
- Probability of two or three photon is proportional to the respectively, fourth and sixth power electric field i.e square and cube of the intensity of radiation field  $(I^n)$ .

6. In general,  $\Gamma_{fg}^{(n)} \infty \sigma_n I^n$ , where,  $\sigma_n$  is the generalized cross-section of the *n* photon ionization (Chin *et al.* 1984).

### In the case for two photon ionization for laser radiation containing different frequency

# For two fields $(\omega_k, \omega_t)$

The same final continuum state is reached by:

- the absorption of two identical laser photons (ω<sub>k</sub>) or (ω<sub>j</sub>) with the polarization vectors ε̂<sub>k</sub>.
- the absorption of two different laser photons (ω<sub>k</sub> andω<sub>j</sub>) with different polarization vectors ε̂<sub>k</sub> and ε̄<sub>i</sub> respectively,
- the absorption of two different laser photons(ω<sub>j</sub> and ω<sub>k</sub>) with different polarization vectors ĉ<sub>j</sub> and ĉ<sub>k</sub> respectively.
- the absorption of two identical laser photons(ω<sub>i</sub>) or (ω<sub>k</sub>) with ε̂<sub>i</sub>.

#### For three fields:

The same final continuum state is reached by the interference of six different routes connected to the absorption of two identical laser photons with polarization vectors ( $\hat{\varepsilon}_k$  and  $\hat{\varepsilon}_j$ ) and absorption of two laser photons with different polarization vectors.

#### In the case for three photon ionization the laser radiation containing different frequency

For two fields The same final continuum state is reached

by the interference of three different routes connected to the absorption of three identical laser photons with polarization vectors  $(\hat{\varepsilon}_l, \hat{\varepsilon}_k \text{ and } \hat{\varepsilon}_i)$ .

by the interference of six different routes connected to the absorption of three different laser photons with polarization vectors( $\hat{\varepsilon}_i$ ,  $\hat{\varepsilon}_k$  and  $\hat{\varepsilon}_l$ ).

#### For three fields:

The same final continuum state is reached

by the interference of nine different routes connected to absorption of three identical laser beams with the polarization vectors  $(\hat{e_i}, \hat{e_k})$  and  $\hat{e_l}$  respectively.

by the interference of eight different routes connected to absorption of three different laser beams with the polarization vectors  $(\hat{\varepsilon}_j, \hat{\varepsilon}_k \text{ and } \hat{\varepsilon}_l)$  respectively.

by the interference of ten different routes connected to absorption of three different laser beams with different polarization vectors( $\hat{e_j}$ ,  $\hat{e_k}$  and  $\hat{e_l}$ ) respectively.

Thus the same final continuum state is reached in total by the interference of twenty seven different routes connected to the absorption of three identical or different laser photons with polarization vectors( $\hat{\varepsilon}_j$ ,  $\hat{\varepsilon}_k$  and  $\hat{\varepsilon}_l$ ).

Besides the energy conservation the total angular momentum of the system (Atom + Photon) has to be conserved. The transition probability therefore depends on polarization of the absorbed or emitted electromagnetic radiation.

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