Photon Assisted Ionization of Atoms

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The atom's ionization rates by two photons is reported for different approaches: semi-classical regime using rate-equations, and quantum optics approach using density matrix operators. For each trend, the assumptions are presented and the ionization rates are given for different special cases like ionizing lasers with Lorenzian and Gaussian beam profiles.

Outline

1. Rate-equation approach

To find the probability of ionization by two or multi photons we consider the following model (Fig. 1) and the assumptions [1]:

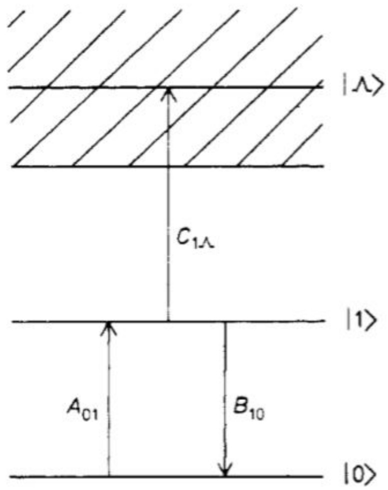

Fig. 1: Model for two photon ionization (Fig. 1 of [1]): |a> are atomic eigenvectors:(|0>: ground state, |1>: first excited state, |Λ>: atomic continuum states (=K,σ) with K as photo-electron wavevector and σ as its polarization, $C_{1\lambda}$ is the rate of ionizing transition from |1> to |λ>, A_{01} is the rate of transitions from |0> to |1>, and B_{10} is the rate of transitions from |1> to |0>.

Assumption

1. The rate of transitions from the final bound level to the continuum is much smaller than the rate of transitions between any two bound states of the atom that are active in the multiphoton ionization process:

   $C_1 \ll A_{01}, B_{10}$; $C_1 \equiv \Sigma_\Lambda C_{1\Lambda}$, so that the levels can achieve a quasi-steady state among themselves.

By this assumption, we can study a case that the laser field produces a steady-state distribution amongst the bound atomic levels, and that photoionization takes place by a small leakage from the quasi-stationary final bound atomic state.

The rate equations for the occupation probabilities of each atomic level are as following where the coefficients are defined in the caption of Fig. 1:

$$\frac{dp_0}{dt} = -A_{01}p_0 + B_{10}p_1$$

$$\frac{dp_1}{dt} = -(B_{10} + C_1)p_1 + A_{01}p_0$$

$$\sum_{\Lambda} \frac{dp_\Lambda}{dt} = C_1 p_1 \quad \cdot(1)$$

While the last equation is equal to the "ionization rate W_2", the problem reduces to finding the steady-state population (probability) of bound atomic levels (p_0 and p_1) under the influence of applied laser field.

Assumptions

2. Steady-state conditions for states |0>, and |1>, i.e. $dp_0/dt=dp_1/dt=0$,
3. The normalization condition $p_0+p_1=1$,

Ionization rate by two photons (W_2) – general formula with assumptions:

The two-photon ionization rate can be easily found considering the above equations as following:

$$W_2 \equiv \sum_{\Lambda} \frac{dp_\Lambda}{dt} = C_1 p_1 = \frac{A_{01}C_1}{A_{01}+B_{10}}. \quad (2)$$

Where:

$C_1 \equiv \Sigma_\Lambda C_{1\Lambda}$ is the total rate from |1> to any |Λ> state, and $C_{1\lambda}$ is the rate of ionizing transition from |1> to |λ>,

A_{01} is the rate of transitions from |0> to |1>,

B_{10} is the rate of transitions from |1> to |0>, and

p_λ is the occupation probability of (continuum) level |λ>.

In the following we introduce new assumptions and find C_1, A_{01}, and B_{10}, to express Eq. (2) as a function of more intrinsic parameters of the atom-laser beam system.

Assumptions

4. $B_{ba} = A_{ab} + 2\gamma_{ab}$: The relation between the direct and reverse transition rates between levels |0> and |1> (as shown in Fig. 1 (A_{01}, and B_{10}, respectively), or any transition rates between any two general states |a> and |b>; where γ_{ab} is the spontaneous transition rate from upper level |a> to the lower level |b>.

5. $\Gamma_a t \ll 1,\ \Gamma_b t \ll 1$; which introduce a time limit that is much smaller than the lifetimes of atomic bound levels |a> and |b> (Γ_i is the inverse of lifetime of state |i> and is equal to the decay rate from level i to any upper or lower state caused by spontaneous emissions or laser field, i.e. $\Gamma_i = \frac{1}{2}\sum_{j<i} B_{ij} + \frac{1}{2}\sum_{j>i} A_{ij}$).

6. Considering a system of units in which: $\hbar = 1$,

7. Stark shifts of atomic energy levels due to the external laser field are considered so that the final energy level is $\bar{E}_i \equiv E_i + S_i$

Assumptions

8. The function $|V_{ab}|^2 \rho(\bar{E}_b)$ is much more slowly varying function of $\bar{E}_b$ than the Lorentzian function of integrand $P_{a,b}(t)$, at resonance $\bar{E}_b = \bar{E}_a$

9. $\Delta E t \gg 1 \gg \Gamma_i t$; where i=a,b and ΔE is the full width at half maximum of $|V_{ab}|^2 \rho(\bar{E}_b)$ with respect to variable $\bar{E}_b$,

10. Considering the energy flux carried by photons at frequency ω as: $$J(\omega) = \int \frac{\omega^3 n(\omega)\, d\Omega}{8\pi^3 c^2}$$, where **Ω** denotes the solid angle of the photon beam and n(ω) is the number of photons in the beam at frequency ω.

11. $n(\omega) \gg 1$; assuming intense laser beams in which the number of photons in the beam at frequency ω are large.

12. The total energy flux carried by photon field is as following:

$$J = \int_0^\infty J(\omega)\, \mathrm{d}\omega \quad (3)$$

For Lorentzian lineshape:

$$J_{\mathrm{L}}(\omega) = \frac{J\Delta/\pi}{(\omega - \omega_{\mathrm{L}})^2 + \Delta^2} \quad (4)$$

Gaussian lineshape:

$$J_{\mathrm{G}}(\omega) = \frac{J}{\sigma\sqrt{2\pi}} \exp\left(-\frac{(\omega - \omega_{\mathrm{L}})^2}{2\sigma^2}\right) \quad (5)$$

The central laser frequency is ω_L and the half width at half maximum Δ for Lorentzian shape and $\sigma(2\ln 2)^{1/2}$ for Gaussian shape.

To find the ionization rate, we consider the probability of transition from a bound state |b> to the another bound one |a> as [1]:

$$P_{a,b}(t) = \frac{|V_{ab}|^2}{\bar{E}^2_{a,b} + \Gamma^2_{a,b}} (\mathrm{e}^{-2\Gamma_a t} + \mathrm{e}^{-2\Gamma_b t} - 2\,\mathrm{e}^{-(\Gamma_a + \Gamma_b)t} \cos \bar{E}_{a,b} t)$$

(Eq. 10 of [1]) which is a Lorentzian function of $\bar{E}_{a,b}$ with decaying rates of levels |a>, |b>, and the superposition of them.

where:

$$V = \sum_\lambda \sum_{i,j} \mathrm{i} \left(\frac{2\pi\alpha\omega_\lambda c}{V}\right)^{1/2} (r_{ij} a_\lambda - r^*_{ij} a^\dagger_\lambda) |i\rangle\langle j|$$

is the interaction potential of the atom with the radiation field in dipole approximation, V is the volume of the system, ω_λ is the frequency, $\boldsymbol{\alpha}$ is the fine structure, a_λ is the annihilation operator for a photon in mode ,r_{ij} is the dipole matrix element and |i> and |j> label atomic states.

Considering |b> as one of the continuum final states,

$$P^b_a(t) \equiv \sum_b P_{a,b}(t) = \int_0^\infty \rho(\bar{E}_b)\, \mathrm{d}\bar{E}_b\, P_{a,b}(t)$$

, where $\rho(E_b)$ is the density of final states,

By above assumptions one yields the Fermi golden rule:

$$P^b_a(t) = (2\pi |V_{ab}|^2 \rho(\bar{E}_b))_{\bar{E}_b = \bar{E}_a} t$$

The transition rates could be found by following relations:

$$\frac{\mathrm{d}P_a^b}{\mathrm{d}t} \equiv A_{ab}$$, and $$\sum_{\Lambda} \frac{\mathrm{d}P_a^{\Lambda}}{\mathrm{d}t} \equiv C_a$$, and assumption 4 ($B_{ba} = A_{ab} + 2\gamma_{ab}$)

considering |a> = |1>, and |b>=|Λ>, the above relations yield: C_1, A_{10}, and B_{01}

Ionization rate by two photons (W_2) for Lorentzian laser beam– including additional assumptions

Considering the above assumptions for the range of validity, one finds the ionization rate of an atom, (schemed in Fig. 1 by two photons), with Lorentzian laser lineshape (Eq. 4):

$$W_2 = \frac{\Omega_{10}^2 C_1 \Delta}{4\gamma_1[\delta_{10}^2 + \Delta^2 + (\Omega_{10}^2 \Delta / 2\gamma_1)]}. \quad (6)$$

where

- C_1 is the total rate from |1> to any |Λ> state,
- Δ is the half width at half maximum of the Lorentzian lineshape of laser beam (Eq. 4),
- δ_{10} is the detuning between the central laser frequency and the energy gap between atomic bound levels |0> and |1>, i.e. $\delta_{10} = E_{10} - \omega_L$,
- Ω^2_{10} is the integrated Rabi frequency for transition |0> => |1> , which in general form is defined as $\Omega_{ab}^2 \equiv 8\pi\alpha |r_{ab}|^2 J$:where r_{ab} is the dipole matrix element of atom regarding two states |a> and |b>, and J is the total energy flux of laser field.
- γ_1 is the spontaneous decay rate from level |1>.

Special case: high power laser beam

For high power laser beams with Lorentzian profile, the condition: $\Omega_{10}^2 \Delta \gg 2\gamma_1(\delta_{10}^2 + \Delta^2)$ is applied [1] in which the parameter $\Omega_{ab}^2 \equiv 8\pi\alpha |r_{ab}|^2 J$ includes ***J*** or the total energy flux carried by photons of laser beam (Eq. 3). In such condition the population of levels |0> and |1> become equal ($p_0=p_1=1/2$) and the ionization rate yields:

$$W_2 \to \tfrac{1}{2} C_1 \quad (7)$$

Ionization rate by two photons (W_2) for Gaussian laser beam– including additional assumptions

Considering the above assumptions, one finds the ionization rate of an atom schemed in Fig. 1 by two photons of a laser beam with Gaussian profile (Eq. 5) as following:

$$W_2 = \tfrac{1}{2}\Gamma^2 C_1[\exp(\delta_{10}^2/2\sigma^2) + \Gamma^2]^{-1} \quad (8)$$

where:

$$\Gamma^2 \equiv (\tfrac{1}{2}\pi)^{1/2} \frac{\Omega_{10}^2}{2\gamma_1\sigma}$$

General solution for rate equation approach

In the following we do not consider assumption 1 ($C_1 \ll A_{10}, B_{01}$). By generally solving rate equations via Laplace transform, one achieves $p_i(t)$; the probability of the atom being in state |i>.

Assumption

1. Initial condition: $p_0(0)=1$, $p_1(0)=0$.

Ionization rate by two photons (W_2) – general formula with NO assumptions:

The ionization rate for the above initial condition and no other previous assumptions is:

$$W_2 \equiv \sum_{\Lambda} \frac{\mathrm{d}p_{\Lambda}}{\mathrm{d}t}$$

where $p_{\lambda}(t)$ is the probability of the atom being in a continuum state. By considering $p_c(t) = \Sigma_{\Lambda}\, p_{1\Lambda}(t)$, the Laplace transform yields:

$$p_c(t) = \frac{A_{01}C_1}{\mu}\left(\frac{1-e^{-\frac{1}{2}(\lambda-\mu)t}}{\frac{1}{2}(\lambda-\mu)} - \frac{1-e^{-\frac{1}{2}(\lambda+\mu)t}}{\frac{1}{2}(\lambda+\mu)}\right) \quad (7)$$

where

$\lambda = (A_{01} + B_{10} + C_1)$, and

$$\mu = [(A_{01} + B_{10} + C_1)^2 - 4A_{01}C_1]^{1/2}$$

Special cases: Long time limit

The probability of the atom being in an ionized state in long time limit is 1, which is obvious for our model, since we have not considered any reverse transition from ionized states to bound atomic states |0> and |1>.

$$\lim_{t \to \infty} p_c(t) = 1 \quad (8)$$

Special cases: Short time limit

The time dependent probability of the atom being in an ionized state in short time limit is proportional to t^2, which is seems not be accurate. It will be shown by density matrix approach (as well as some other works referred to in [1]) that in short time limit the ionization probability is proportional to t^3.

$$\lim_{t \to 0} p_c(t) = \tfrac{1}{2} A_{01} C_1 t^2 \quad (9)$$

Special cases: time-independent ionization rate

For time intervals in the following regime:

$$\frac{2}{|\lambda - \mu|} \gg t \gg \frac{2}{|\lambda + \mu|}$$

which is a necessary condition for the relation:

$$(A_{01} + B_{10} + C_1)^2 \gg 4A_{01}C_1$$

which is again a necessary condition for assumption 1 ($C_1 \ll A_{10}, B_{01}$).

the ionization rate is time-independent and is found as:

$$W_2 = A_{01} C_1 / \mu \quad (10)$$

where λ and μ are defined in the previous case.

Ionization rate for the time-independent regime with Lorentzian lineshape

With the above condition for transition rates, and assuming a Lorentzian lineshape of laser as Eq. (4), the ionization rate is as following:

$$W_2 = \frac{\Omega_{10}^2 C_1 \Delta}{4(\gamma_1 + \frac{1}{2}C_1)[\![\delta_{10}^2 + \Delta^2 + \{\Omega_{10}^2 \Delta / [2(\gamma_1 + \frac{1}{2}C_1)]\}]\!]} \quad (11)$$

which differs from Eq. (6) by that γ_1 is replaced by $\gamma_1 + \frac{1}{2}C_1$.

2. Quantum optics approach

2.1. Density matrix approach

The rate equation approach is not valid when laser field is well described as a single mode, since assumption 9 of previous section ($\Delta E t \gg 1 \gg \Gamma_i t$) is not true for any time t. However we can assume that in some situations, the two-level system comes into a quasi-steady state with itself before appreciable loss to the continuum can occur ($dp_i/dt=0$, where p_i is the probability of atom being in the state |i>).

In the density matrix approach, equation of motion are given by following master equation

$$\frac{\partial \rho}{\partial t}+\mathrm{i}[H,\rho]-\gamma_1(2|0\rangle\langle 1|\rho|1\rangle\langle 0|-\rho|1\rangle\langle 1|-|1\rangle\langle 1|\rho)=0 \tag{2.1}$$

[Agarwal 1974], in RWA (rotating wave approximation) Hamiltonian:

$$H=a^{\dagger}a\omega+\omega_0|1\rangle\langle 1|+ga|1\rangle\langle 0|+g^*a^{\dagger}|0\rangle\langle 1|. \tag{2.2}$$

where a, a^+ are the annihilation and creation operators of the single mode EM field with frequency w, w0 is the energy difference between two atomic levels $\omega_0=E_{0,1}$, g is the coupling constant and is related to Rabi frequency by $\Omega_{10}^2=4|g|^2 n$

Assumptions

1. the coupling constant g is considered real for simplicity,
2. The laser beam is intense so that the number of photons in frequency ω is large: $n(\omega)>>1$

Ionization rate- general form

By solving the equations of motion obtains from master equation (2.1), and above assumptions [1], the ionization rate is found as:

$$W_2\equiv\rho_{n1;n1}C_1=\tfrac{1}{4}\Omega_{10}^2C_1/(\delta_{10}^2+\gamma_1^2+\tfrac{1}{2}\Omega_{10}^2) \tag{2.3}$$

where:

- $\rho_{n1;n1} = \frac{1}{4}\Omega_{10}^2/(\delta_{10}^2 + \gamma_1^2 + \frac{1}{2}\Omega_{10}^2)$,

  ($\rho_{n1;n1} = \langle n|\langle 1|\rho|1\rangle|n\rangle$, and the reduced density matrix for atom alone is: $\rho_{jj} = \sum_n \rho_{nj;nj}$, where i=0,1 is the atomic levels, and n is the photon number),

- $\delta_{10} \equiv \omega_0 - \omega$

Eq. (2.3) differs from ionization rate of Eq. (6) obtained by rate-equation approach, by setting $\Delta = \gamma_1$.

Special case: Short time limit

considering the initial condition:

$\rho_{n+10;n+10}(0) = 1$ and all other matrix elements of $\rho(0)$ being zero, the probability of the atom being in an ionized state would be:

$$p_c \equiv \sum_\Lambda \rho_{n-1\Lambda;n-1\Lambda}(t) = \tfrac{1}{12}\Omega_{10}^2 C_1 t^3 \quad (2.4)$$

Special cases: Inclusion of transitions to the continuum states

If not considering the steady-state model anymore, and take into the account a non-negligible rate of transitions from state |1> to the continuum states, in an intuitive way by adding two corresponding coefficients rates to the equations of motions [1], one finds that for the following parameter regime:

$$\frac{\Omega_{10}^2 C_1}{4\kappa} \ll \kappa^2 + \delta_{10}^2 + \tfrac{1}{2}\Omega_{10}^2\left(1 + \frac{C_1}{2\kappa}\right),$$

the ionization probability is:

$$W_2 = \frac{\frac{1}{4}\Omega_{10}^2 C_1}{\delta_{10}^2 + \kappa^2 + \frac{1}{2}\Omega_{10}^2} \quad (2.5)$$

where $\kappa = \gamma + \frac{1}{2}C_1$.

Eq. (2.5) differs from Eq. (2.3) in that γ has been replaced by κ.

Multimode laser beam

If the laser won't be single mode as in the previous section, and the ionizing laser has a finite bandwidth, the density matrix approach would be treated by the following multimode Hamiltonian:

$$H = \sum_{\lambda} a_{\lambda}^{\dagger} a_{\lambda} \omega_{\lambda} + \omega_0 |1\rangle\langle 1| + \sum_{\lambda} g_{\lambda} (a_{\lambda} |1\rangle\langle 0| + a_{\lambda}^{\dagger} |0\rangle\langle 1|) \tag{2.6}$$

where λ is the label of different modes in the laser beam.

Assumptions

1. considering rotating wave approximation (RWA),
2. coupling constant g_{λ} be real for simplicity,
3. The initial state of the field would be a Fock state $|n_1 n_2 n_3 \ldots\rangle \equiv |\boldsymbol{n}\rangle$, where n_i is the number of photons in mode λ_i,

   the state $|\boldsymbol{n} \pm \lambda_k\rangle \equiv |n_1 n_2 \ldots (n_k \pm 1) \ldots\rangle$ is identical to the initial state except that a photon in mode k has been added or removed,
4. The reduced density matrix for atom alone is: $\rho_{jj} = \sum_{\boldsymbol{n}} \rho_{\boldsymbol{n}j;\boldsymbol{n}j}$, where i=0,1 is the atomic levels, and n is the photon number
5. $n_{\lambda} \gg 1$ for all modes λ or equivalently $\rho_{\boldsymbol{n}-\lambda 1;\boldsymbol{n}-\lambda 1} \simeq \rho_{\boldsymbol{n}1;\boldsymbol{n}1}$,
6. Normalization condition: $\rho_{00} + \rho_{11} = 1$ for the reduced density matrix of atom,
7. The diagonal elements ($\rho_{\boldsymbol{n}-\lambda 1;\boldsymbol{n}-\lambda 1}$ and $\rho_{\boldsymbol{n}0;\boldsymbol{n}0}$) are much larger than the off-diagonal elements while they have higher orders of coupling constant g.

Ionization rate - multimode laser beam

Solving the master equation (2.1) for multimode Hamiltonian (2.6), the steady state case of $\partial/\partial t = 0$ for the corresponding density matrix element could be obtained (ρ_{11}). The ionization rate for such case is:

$$W_2 = \rho_{11} C_1 = \frac{2\pi\alpha |r_{01}|^2 C_1 \mathcal{J}}{1 + 4\pi\alpha |r_{01}|^2 \mathcal{J}} \quad (2.6)$$

where

$$\mathcal{J} = \int \frac{J(\omega)\, \mathrm{d}\omega}{(\omega - \omega_0)^2 + \gamma_1^2}$$

for any arbitrary energy flux function J.

Ionization rate by density matrix approach for multimode laser beam- Gaussian lineshape

$$W_2 = \tfrac{1}{2}\Gamma^2 C_1 [\exp(\delta_{10}^2 / 2\sigma^2) + \Gamma^2]^{-1} \quad (2.7)$$

where

$$\Gamma^2 \equiv (\tfrac{1}{2}\pi)^{1/2} \frac{\Omega_{10}^2}{2\gamma_1 \sigma}$$

which is identical to that resulted by rate-equation approach (Eq. (8)).

2.2. Resolvent formalism

The Hamiltonian of the system of atom-laser field is as following:

$$H=\sum_{\lambda} a_{\lambda}^{\dagger}a_{\lambda}\omega_{\lambda}+\sum_{a=0}^{\infty}|a\rangle E_a\langle a|+\sum_{\lambda}\sum_{a,b}(g_{ab}^{\lambda}|a\rangle\langle b|a_{\lambda}+\mathrm{HC}) \qquad (1)$$

The Hamiltonian parameters are ($\hbar$=1) [3]:

- $a_{\lambda}^{\dagger}$, and a_{λ}: creation and annihilation operators of a photon of mode λ (emitted photoelectron) with polarization index η, wave vector **k**, and energy ω_{λ} ,So $\lambda\equiv(\eta, \mathbf{k}, \omega_{\lambda})$, (the notation $n_{\lambda}=a_{\lambda}^{\dagger}a_{\lambda}$ will be used later.)
- E_a: atomic eigenenergies
- |a>: atomic eigenvectors (|0>: ground state, |1>: first excited state, |2> second excited state, |Λ>: atomic continuum states (=K,σ) with K as photo-electron wavevector and σ as its polarisation.
- g^{λ}_{ij}: coupling constant given in the dipole approximation,

$$g_{ab}^{\lambda}=\mathrm{i}(2\pi\alpha\omega_{\lambda}c/V)^{1/2}\langle a|\hat{e}_{\lambda}\cdot\mathbf{r}|b\rangle$$

α : fine structure constant which is $e^2/4\pi\varepsilon_0\hbar c$

$\mathbf{e}_{\lambda}$: unit polarization vector of the field in mode λ,

r: The dipole moment operator of atom divided by electron charge *e*,

V: the volume of the system exposed to laser beam.

- $|\mathbf{n}\rangle = |n_{\lambda1}, n_{\lambda2}, n_{\lambda3}, \ldots\rangle$: Initial laser field state, Fock state with $n_{\lambda i}$ as the number of photons in wavelength λ_i
- $|\mathbf{n}, \mp\lambda_i\rangle = |\mp\lambda_i\rangle = |n_{\lambda1}, n_{\lambda2}, \ldots, n_{\lambda i}\mp 1, \ldots\rangle$: Field states in which a photon in one mode is absorbed or emitted:
- $|a\rangle|\mathbf{n}\rangle = |i, \mathbf{n}\rangle$, or $|i, \mathbf{n}\mp\lambda_i\rangle$: Unperturbed atom-field state:
-

$$|0,n\rangle \begin{matrix}\nearrow & |a,-\lambda_1\rangle & \searrow \\ \searrow & |a,-\lambda_2\rangle & \nearrow\end{matrix} |\Lambda,-\lambda_1-\lambda_2\rangle$$

Evolution of the system by resolvent formalism

Using resolvent formalism [3], the unitary evolution operator of atom-field system could be found using the resolvent function G as following. In other words, the matrix elements of the time evolution operator at time t can be obtained by the following inversion formula:

$$U_{\Lambda,0}(t) = -\frac{1}{2\pi i}\oint dz\ e^{-izt} G_{\Lambda,0}(z) \tag{2}$$

where resolvent function G(z) = 1/(z-H), H is the atom-field system Hamiltonian defined in Eq. (1), and z is the complex variable.

Ionization probability by two photons ($P_2(t)$) – general formula

Two photon ionization probability in a time interval *t* from initial state |0> to final continuous state |Λ> is $P_2(t)$ formulated as following:

$$P_2(t) = \sum_{\Lambda} \mathrm{Tr}(\hat{\rho}_F U_{\Lambda,0}(t) U^{\dagger}_{0,\Lambda}(t)) \tag{3}$$

Where ρ_F is the density matrix of the field, and U is the unitary evolution operator of the system from state |0> to |Λ>.

Assumptions and approximations to calculate $P_2(t)$ by above equation:

1. Considering density matrix of the field as a pure Fock state of $|n_1n_1n_3...\rangle\langle n_1n_2n_3...|$.
2. Γ_i is defined as the inverse lifetime of level |i> due to the spontaneous emission and the effect of perturbation caused by laser field, then:
   Γ_λ=0 means that ionization process is irreversible. So once ion formed, it is perfectly stable and recombination is neglected. This is typical in multiphoton ionization experiments performed at very low gas pressures.
3. Change of sum to integral over all polarization states, wave vectors:
   $$\sum_{\Lambda} \rightarrow \sum_{\sigma} \int d\Omega_{\boldsymbol{K}} \int_{E_I}^{\infty} dE_{\boldsymbol{K}} \rho(E_{\boldsymbol{K}})$$
   where Ω_k is the propagation direction of the λ^{th} mode,
   $\rho(E_{\boldsymbol{K}}) = Vm(2mE_{\boldsymbol{K},I})^{1/2}/8\pi^3$ is the density of final states for the photoelectron,
   $E_{k,I}$ is the ionization energy.

To calculate the integral over all ionization energies, we change the lower limit of integral from E_I to -∞ which is justified well above threshold.

4. The laser beam spectrum I(ω) is defined as following:

$$J(\omega)=\frac{J\Delta/\pi}{(\omega-\omega_0)^2+\Delta^2}$$

where I(ω)=J(ω)dω is the energy in the field carried by photons. This assumption enables us to treat the resonant case, when the central laser frequency is close to an atomic transition frequency. In the ordinary perturbation treatment the denominator of this **Lorentzian distribution** with respect to ω_0 will vanish and simplifies the solution.

5. In addition, the laser has a bandwidth sufficiently large for us to ignore the Stark splitting of the atomic energy levels, so that the laser produces just shifts and broadenings.

Ionization probability by two photons ($P_2(t)$) – including more assumptions

Considering the above assumptions, the following formula is the probability of ionizing the atom by two photons at time t ($P_2(t)$):

$$P_2(t)=\left(\frac{2\pi\alpha c}{V}\right)^2\sum_{a,b}R_aR_b^*\left(\sum_{\lambda_1,\lambda_2}n_1n_2\omega_1\omega_2(f_{1,1}(t)+f_{1,2}(t))-\sum_{\lambda_1}n_1^2\omega_1^2f_{1,1}(t)\right) \tag{4}$$

where n_1, n_2, ω_1, and ω_2, are denoting the number and energy of photons in λ_1 and λ_2 modes, respectively,

$$R_a\equiv r_{0a}r'_{a\Lambda} \quad \text{where: } \sum_\sigma\int d\Omega_{\boldsymbol{K}}r_{a\Lambda}r^*_{b\Lambda}\rho(E_0+2\omega_0)\equiv r'_{a\Lambda}(r'_{b\Lambda})^*$$

Special case 1: $P_2(t)$ via single mode laser field

In this case the number of photons in all modes are zero except one as following:

$$n_\lambda=n_0\delta_{\lambda\lambda_0}$$

It could be shown that in this case, the general form of AIP-TP reduces to the following form:

$$P_2(t)=F^2\sum_{a,b}R_aR_b^*f_{0,0}(t) \tag{5}$$

where R is defined for Eq. (4)

where F= $2\pi\alpha J$ and J=$n_0\,\omega_0\,c/V$ is the energy that is crossing unit area per second.

Special case 2: $P_2(t)$ via continuous mode laser field

Considering the following conversion of sum to integral:

$$\sum_{\lambda} \rightarrow \frac{V}{8\pi^3} \sum_{j} \int k^2\,\mathrm{d}k \int \mathrm{d}\Omega_k$$

And considering a Lorentzian shape for the photon occupation number distribution:

$$n_k = \frac{A\Delta_k/\pi}{(k-k_0)^2 + \Delta_k^2}$$

where $2\Delta_k$ is the half width of the photon distribution n_k in k space.

$$P_2(t) = (2\pi\alpha)^2 \int J(\omega_1)\,\mathrm{d}\omega_1 \int J(\omega_2)\,\mathrm{d}\omega_2 (f_{1,2}(t) + f_{1,1}(t)) \tag{6}$$

Where

$$J(\omega) \equiv I(\omega)\omega = \frac{\omega^3}{8\pi^3 c^2} \int n(\omega)\,\mathrm{d}\Omega$$

<The other parameters need to be investigated in more details as their general definitions are not defined in the corresponding papers.>

References

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