Interference effects on the probe absorption in a driven three-level atomic system by a coherent pumping field

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Abstract. In this work the quantum interference effect on the weak probe absorption in a closed three-level *V* atomic system driven by a coherent driving field is shown to result from the two-quantum processes, constructive for the amplification channels and destructive for the absorption channel. A fourth state in the atom is coupled incoherently to the *V* system and acts as both an incoherent pumping reservoir and as a stationary, final state in perturbation theory. The application refers to Be-like carbon ions where non-linear pump process of the $1s^22s5s(^1S^e)$ - $1s^22p5s(^1P^0)$ transition is combined with the absorption or emission of a single probe photon corresponding to $1s^22s5s(^1S^e)$ -$1s^22p7s(^1P^0)$ transition. The system is analyzed within the *R*-matrix Floquet theory and codes results.

Keywords: laser-induced or assisted processes, atoms in laser field, electromagnetically induced transparency

Introduction

Doubly excited states with two electrons are the most fundamental atomic systems which autoionize. Atoms with two valence electrons offer several advantages for experiments on ultra-cold Rydberg gases and plasmas. Narrow intercombination lines allow extremely high spectroscopic resolution. Recently, it was shown [1] that the electromagnetically induced transparency (EIT) phenomenon can be used to probe Rydberg states of rubidium atoms in a vapor cell non-destructively, and with high resolution. In combination with strong Rydberg - Rydberg interactions, this could also be used to realize a phase gate between single photons [2]. Although EIT was first observed in strontium [3] most subsequent work was focused on the alkali metals. EIT can render resonant transitions transparent, and can open the door to highly efficient XUV and X-Ray Laser Without Inversion (LWI) lasers operating between the ground state and first excited state [4- 7]. Essentially it breaks the symmetry between absorption and stimulated emission by using a laser whose frequency closely matches the transition between the upper level and some other auxiliary level. The lasing medium can be coherently prepared in such a way that absorption between the lower and upper levels is reduced or eliminated. The theoretical mechanism for this process is quantum mechanical interference between two different paths leading to the excited state. The idea is to choose the atom-laser quantum system in such a way that opposite phases are associated to the individual amplitudes, resulting in a destructive interference, which implies that the total probability to make a transition from the lower level to the upper level is also zero. With a zero probability of absorption, the quantum system is transparent to the radiation of the upper - auxiliary levels transition.

Quantum interference between two laser-induced channels can lead to the elimination of the spectral line at the driving laser frequency in the spontaneous emission spectrum, and to the existence of a dark line in the spontaneous emission from one of the excited sublevels. In this work, the basic system consists of a *V*-type atom, with a single "ground" Rydberg excited state and two, closely spaced, autoionizing Rydberg states, damped (or not) by the usual vacuum interactions, so that the two decay pathways from the autoionizing Rydberg states to the excited Rydberg 'ground' state are not independent.

The *V* system requires both *incoherent pumping* and *coherent pumping* in order to have optical gain. A fourth state in the atom is coupled incoherently to the *V* system and acts as both an incoherent pumping reservoir and as a stationary, final state in perturbation theory. The paper is structured as follows. In Section 2 we briefly outline the theory and the details of calculation. The calculation reported here is part of a general investigation which started with studies of $\Delta n = n'\text{-}n = 0, 2$, n, n' ranging from 5 to 12 in C^{2+}. The laser induced degenerate states phenomenon has been studied for $1s^2 2pns\ (^1P^0) \rightarrow 1s^2 2sn's(^1S^e)$ transitions in C^{2+} [8,9]. We only recall here the principal ideas of the theory and the numerical results. One interesting feature of the present calculation is to show the role of the laser-induced degenerate state phenomenon (LIDS) in selecting the resonances of the dressed atom-field system. Only the relevant equations will be repeated here. Energy level and pumping scheme for the four-level atom corresponding to C III ion is shown in Figure 1. Following our previous works we assume the laser beams to be purely monochromatic, linearly polarized, and spatially homogeneous. The details of the calculation approach are spelled out in this section. It is analyzed the situation when two excited Rydberg states are resonantly coupled by an intense, monochromatic, monomode, linearly polarized laser field. In order to understand the general features of these structures embedded by the field, we have used a model that retains the essential ingredients of the full *R*-matrix Floquet [10] calculation, namely, a bound state coupled nonperturbatively by the field to an autoionizing state and to the continuum. In this section we give the density-matrix results for this system. In particular , for $\delta = \delta'$, where δ,δ' are the pump and probe detunings from the atomic resonances, respectively, no population inversion on the probe transition is possible when the bare, upper probe level decays faster than the bare, upper pump level. Nevertheless, probe amplification can occur under these conditions. Finally in Section 3 we draw conclusions from this work and we discuss future directions of research.

2. Theory and details of calculations

In general one associates double-resonant processes with processes induced by two lasers. In our case the 'two beams' are the laser beam and the electron beam. Thus, instead of using a probe laser to reach a high-lying state in the continuum, this state can be reached by choosing the energy of the incident electrons to be at resonance with the resonant state of the composite electron-atom system. An intense laser is then used to strongly couple the autoionizing state to another state. The advantage of such approach is that it allows flexibility: tuning the energy of the electrons over a wider energy range is easy, while lasers operate in restricted frequency intervals. Additionally, resonant states of all symmetries can be excited, not only the dipole selected states.

Our description of the laser field assumes that the following conditions hold: (i) the number of photons per mode (in our case there is only one mode) of the laser is large so that the field may be treated classically; (ii) the wavelength of the laser is large compared with the spatial extend of the atom so that the dipole approximation is applicable; and (iii) the pulse duration is long compared to typical scattering times.In our *V* atomic system, the two 1 and 3 states (see Figure 1) are autoionizing states for C^{2+} , while with 2 is labeled a highly excited Rydberg state. We recall here that the $1s^22p7s$ ($^1P^0$) – $1s^22s5s$ ($^1S^0$), corresponding to 3 -2 transition in Figure 1(*a*), is a radiative transition as has been demonstrated in Ref. 9.

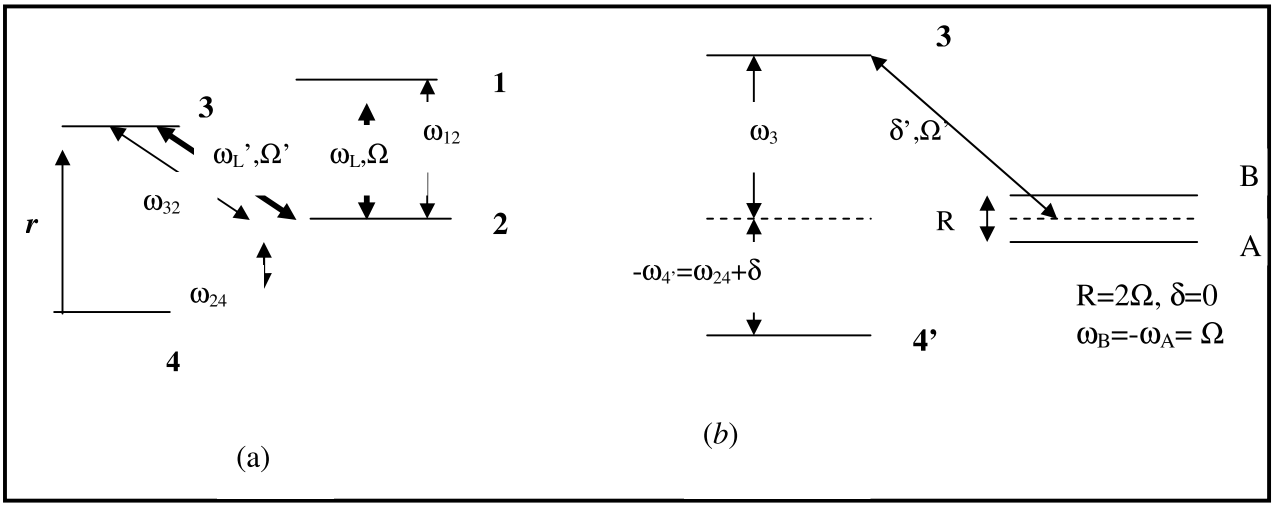


Figure 1. (*a*) Energy level and pumping scheme for the four-level CIII atom system, in which *r* is the incoherent pumping rate of level 3, here $1s^22p7s(^1P^0)$ level. The coherent pump drives the $1s^22p5s$ $(^1P^0)$ $-1s^22s5s(^1S^e)$, i.e. 1-2 transition strongly with a Rabi frequency Ω. Amplification without inversion occurs for the probe laser on the 2 -3 , i.e. $1s^22s5s(^1S^e)$ – $1s^22p7s(^1P^0)$, transition. State 4, i.e. $1s^22s^2$ state, is asymptotically stable. (*b*) Dressed energy levels of the atom-strong pump field system for $\delta = \omega_L - \omega_{12} = 0$ in the frame rotating at pump frequency ω_L. The probe field at frequency ω_L' and the quantum electromagnetic field (not shown) cause transitions between state $1s^22p7s(^1P^0)$ (labeled as 3) and the dressed states A and B, which are split in energy by the generalized Rabi frequency $\hbar\Re = 2\hbar\Omega$. Interference results when a superposition of dressed states are formed as an intermediate state during probe field scattering. The dashed line marks the zero of energy (LIDS position).

In Table 1 are presented field free energies of these states as taken from Ref.9. We restrict ourselves to the *V* atomic system shown in Figure 1: two autoionizing states $1s^22p5s$ and $1s^22p7s$ are coupled by the field to the $1s^22s5s$ Rydberg state and to the continuum. A strong pump laser field couples the $1s^22s5s(^1S^e)$ and $1s^22p5s(^1P^0)$ states. We make use of previously reported results to analyze these states dressed by the field. For certain values of pump field's intensity and frequency, the two quasi-energies become equal, giving rise to laser-induced degenerate states (LIDS).

Table 1. The $1s^2 2sns(^1S^e)$, $1s^2 2pns(^1P^0)$ and $1s^2 2pnd(^1P^0)$ states energy relative to $^2S^e$ and $^2P^e$ ionization thresholds.

n	5	6	7	8	9	10	11	12
2s*n*s	-0.21518	-0.13849	-0.10106	-0.076644	-0.060059	-0.0483114	-0.03967	-0.03315
2p*n*s	0.09376	0.159126	0.197516	0.221925	0.238416	0.25007	0.25861	0.26506
2p*nd*	0.1225	0.17549	0.20769	0.22868	0.24313	0.25350	0.26118	0.26703

In Figure 2 we reproduce the behavior of the real and imaginary parts of the energy belonging to the $1s^2 2s5s(^1S^e)$ and $1s^2 2p5s(^1P^0)$ states dressed by the strong pump field of 1×10^{13} W cm^{-2} intensity and tuning frequency near resonance. The main result of these calculations was as follows. Close to LIDS points, LIDS frequency for fixed intensity, can be predicted as complex trajectories with a real part avoided crossing and an imaginary part crossing switch to having a real part crossing and an imaginary part avoided crossing as ω passes through LIDS frequency. We make use of these primary calculations.

Figure 1(*a*) shows the bare energy-level separations, $\hbar\omega_{12}$ and $\hbar\omega_{32}$, and the laser field frequencies, ω_L and ω_L', for the pump and probe transitions, respectively. The strong pump transition between states 1 and 2 has an associated Rabi frequency Ω. The system is studied in a dressed basis, taking into account the strong laser-atom interaction. Following our previous work, the atomic system is then dressed by a monochromatic, linearly polarized laser field of frequency ω and intensity *I*. The time-dependent Schrödinger equation is solved by expanding the wavefunction as a Floquet-Fourier series. This yields a block structure of the Hamiltonian, each block corresponding to the absorption or emission of a specific number of photons. By diagonalizing the Hamiltonian matrix in the inner region in length gauge, we obtain the R-matrix at the boundary of the box (radius *a*). This matrix, transformed from the length to the velocity gauge, is propagated over an outer region to a radius *a*', where the long range interactions are small. At this radius, we match the R-matrix with an asymptotic expansion of the wave function to obtain the energy E and width Γ of the system as a complex quasi-energy $E_F = E - i\Gamma/2$.

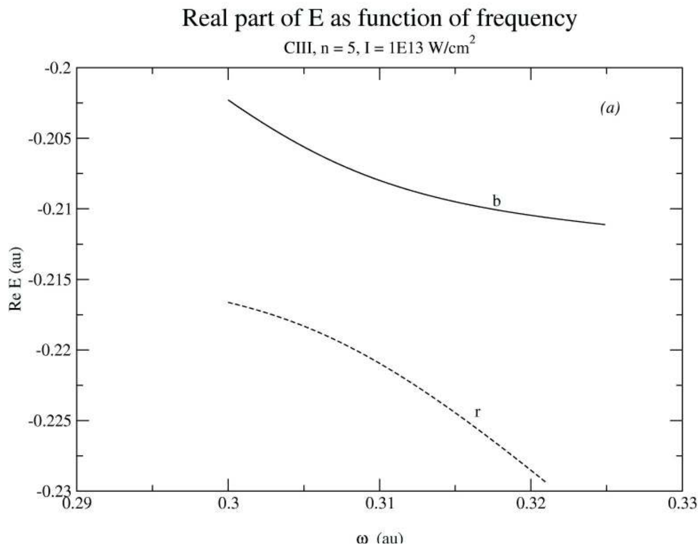


Figure 2. (*a*) . The real part of E as function of frequency, in atomic units, for an intensity of the pump laser 10^{13} W/cm^2 and belonging to $1s^2 2s5s(^1S^e)$(solid line) and $1s^2 2p5s(^1P^0)$ (dashed line) states in CIII. States are labeled by 'b' and 'r' respectively

Dressed energy levels of the atom-strong pump field system for $\delta = \omega_L - \omega_{12} = 0$ are difficult to determine. The LIDS energy can be estimated quite well as soon as the classic LIDS curves are seen in the complex plane even if the frequencies are not very close to the LIDS value. For our strong pump field dressed states this position corresponds to the energy value of $\omega_A = \omega_B = -0.21385$ atomic units (*au*) in Fig 1(*b*). Following our model, if the weak probe field is tuned at frequency $\delta' = \omega_L' - \omega_3$ (Figure 1(*b*)), density-matrix results will show the probe amplification. This amplification is due to quantum interference results when a LIDS state is formed as an intermediate state during probe scattering. Moreover, for $\delta - \omega_L - \Gamma^r = \delta' - \omega'$, with Γ^r radiative transition probability, there is a net probe gain coefficient associated with a population difference of dressed or bare states in the absence of the probe field. Following *density-matrix approach* to the calculation there is no population inversion in either the dressed or bare picture if $\Gamma_3^r / \Gamma_1^r > 1$, where Γ_3^r and Γ_1^r are the state 3 and state 1 decay rates, respectively. As a consequence of this condition, the terms arising from the population difference can result in probe absorption only. On the other hand the ratio Γ_3^r / Γ_1^r can be associated with a coherence of dressed or bare states in the absence of the probe.

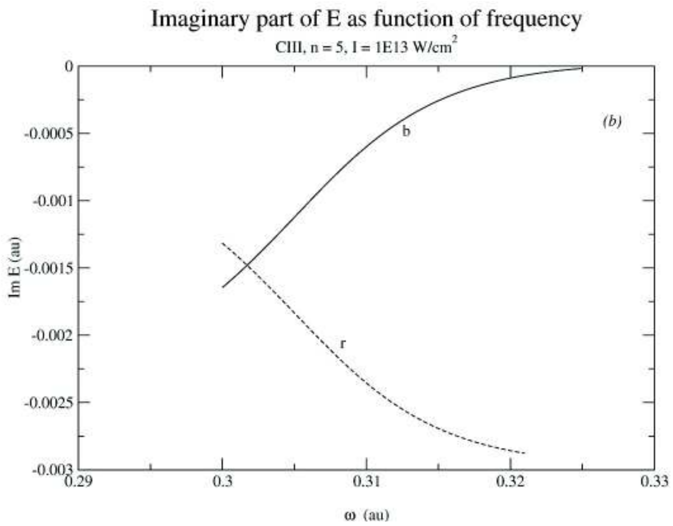


Figure 2. (*b*) The imaginary parts of *E* as function of frequency, in atomic units, and for pump laser intensity of 10^{13} W/cm^2. The curves correspond to $1s^2 2sns(^1S^e)$ and $1s^2 2pns(^1P^0)$ states in C^{2+}, n = 5 and have solid or dashed line, respectively.

First we consider the steady state case, namely $\dot{\rho}(t) = 0$. According to the choice made (Fig. 1 (b)) the Rabi frequencies have the expressions

$$2g = \frac{2}{\hbar}\vec{d}_{13}\cdot\vec{\varepsilon}_1 E_1 = 2c\sqrt{\frac{3\gamma_1\cdot I_1}{\hbar\left|W_{13}\right|^3}}\sin\theta \tag{1}$$

$$2G = \frac{2}{\hbar}\vec{d}_{23}\cdot\vec{\varepsilon}_2\cdot E_2 = 2c\sqrt{\frac{3\gamma_2\cdot I_2}{\hbar\left|W_{23}\right|^3}}\sin\theta \tag{2}$$

where I_1 şi I_2 are the probe field intensity and the coupling field intensity, respectively. We have calculated the density matrix element ρ_{13} for a resonant coupling, $\Delta_2 = 0$, the frequency of coupling laser $\omega_2 = 0{,}3085$ atomic units (au), the intensity of probe laser $I_1=10^{13}$ W/cm^2. Varying the intensity of the coupling laser, for different values of the coupling laser intensities, and polarization angle $\theta = 45°$ we observe a window in the Im(ρ_{13}), that indicates the minimum the probe-laser absorption. This is electromagnetically induced transparency which appears for coupling laser intensities with values from $5\cdot10^{12}$ Wcm^{-2} to 10^{13} Wcm^{-2} (Fig. 3). At the higher intensity of the coupling laser the absorption of the probe laser by the atom is smaller.

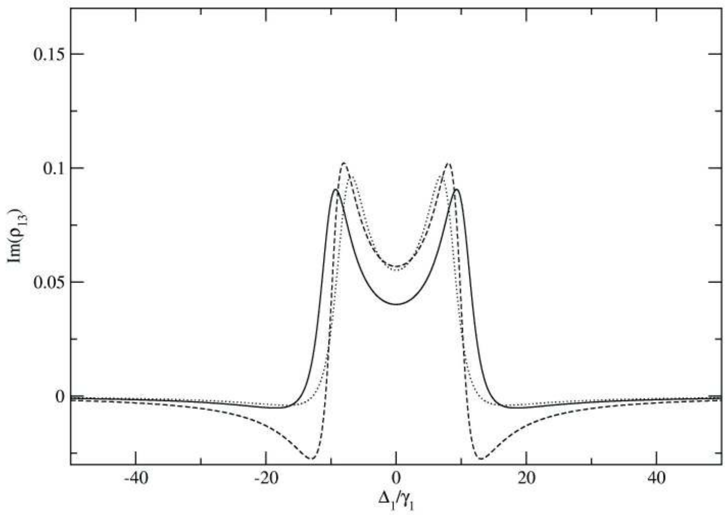
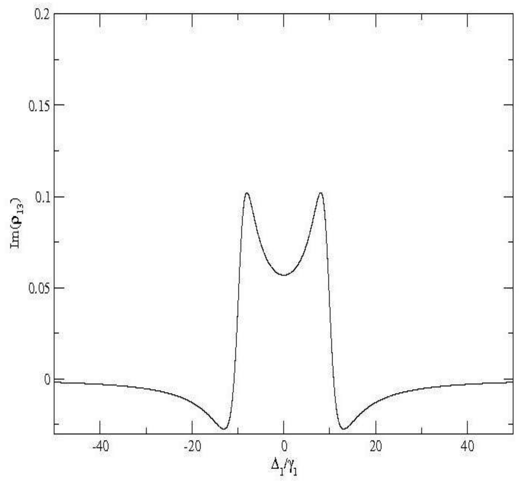
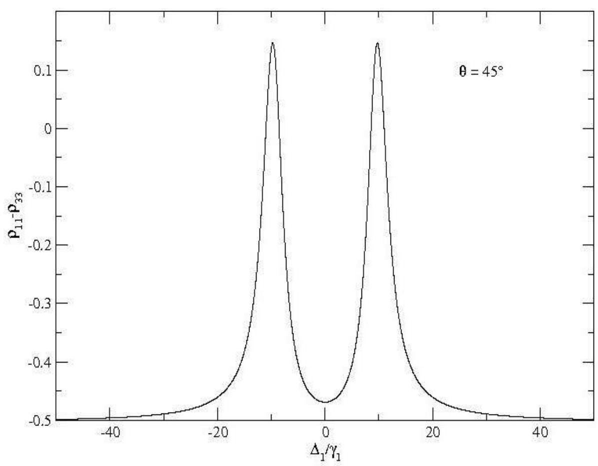


Figure 3. Quantity Im(ρ_{13}) who characterize probe laser absorption for Vee system C^{2+}. The coupling laser intensities are: $I_2=10^{13}$ Wcm^{-2} for the solid curve, $I_2=8\cdot10^{12}$ Wcm^{-2} for the dashed curve and $I_2=5\cdot10^{12}$ Wcm^{-2} for the dotted curve. We take coupling laser frequency $\omega_2=0{,}3085$ au, probe laser intensity $I_1=10^{12}$ Wcm^{-2}, coupling laser detuning $\Delta_2 = 0$, $\theta = 45°$ and parameter of vacuum induced coherence $\eta_0=1$.

From Fig. 3 we observe that at $8\cdot10^{12}$ W/cm^2 there is gain, namely Im(ρ_{13}) < 0. We have calculated the populations gap for the **1s^{2}2p7s(^{1}P^0)** and **1s^{2}2s5s(^{1}S^e)** states, respectively $\rho_{11} - \rho_{33}$ (Fig. 4. (b)), and $\rho_{22} - \rho_{33}$ (Fig. 4. (c)), where ρ_{22} is the population of the **1s^{2}2p5s(^{1}P^0)** state. Calculated values of ratio Δ_1/γ_1 for which $\rho_{11} - \rho_{33}<0$ and $\rho_{22} - \rho_{33}<0$ indicated the gain presence, as well. In this case there is gain without population inversion.



(a)



(b)

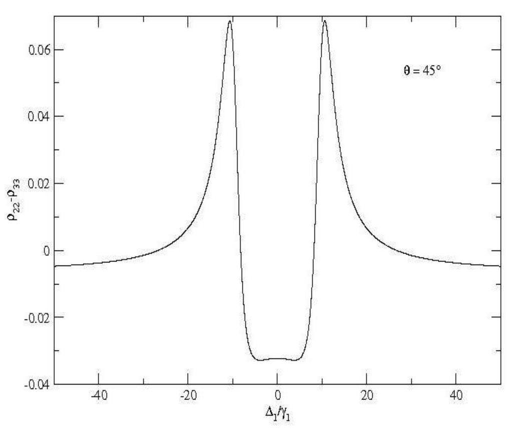


Figure 4. There is gain without population inversion for the coupling laser intensity $I_2 = 8\cdot10^{12}$ Wcm^{-2} and the angle $\theta=45°$ (a) Im(ρ_{13}) who is directly proportional with the absorption coefficient of the probe laser by the medium (b) difference between populations of state 1s^{2}2p7s(^{1}P^0) and the of the state 1s^{2}2s5s(^{1}S^e), $\rho_{11} - \rho_{33}$ and (c) difference between populations of state 1s^{2}2p5s(^{1}P^0) and the of the state 1s^{2}2s5s(^{1}S^e), $\rho_{22} - \rho_{33}$.

3. Conclusions

In this work we have studied the possibility of using the R-matrix Floquet theory and code results from the non-perturbative treatment of laser-induced degenerate state phenomenon in Be-like C ion, and the general formulation of the electromagnetically induced transparency process, to study the role of quantum interferences on the population transfer between highly excited Rydberg state and an autoionizing state. Following our preliminary results, based on the density-matrix equations in dressed-state basis, we conclude that the three-level V system which has been analyzed in this work, cannot exhibit lasing without population inversion if driven by a single field only. Additional pumping has to be applied. Finally, in order to probe the above-calculated quantum interference processes at resonance, we consider a laser pulse train propagating under electromagnetically induced transparency conditions. This work is in progress.

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References:

[1] A. K. Mohapatra, T.R. Jackson, C.S. Adams, Phys. Rev. Lett. **98 ,** 113003 (2007).

[2] I. Friedler, I. Petrosyan, *et al.* Phys. Rev. A **72** 043803 (2005)

[3] K-J Boller, A. Imamolu, S. E. Harris Phys Rev Lett. **66** , 2593 (1991).

[4] A.K. Patnaik, P.S. Hsu, G. S. Agarwal, G.R. Welch, M.O. Scully, Phys. Rev. A**75**, 23807(2007);

[5] V.V. Kozlov, Y. Rostovtsev, M.O. Scully, Phys. Rev. A **74**, 63829(2006),

[6] Y. V. Rostovtsev, Z-E Sariyanni, M.O.Scully, Phys.Rev. Lett. **97,** 11301(2006)

[7] M.G. Bason, A. K. Mohapatra, K.J. Weatherill, C.S. Adams, J. Phys. B: At. Mol. Opt. Phys. **42** 075503 (2009)

[8] V. Stancalie, Physics of Plasmas **12**, 043301 (2005)

[9] V. Stanaclie, Physics of Plasmas **12**, 10075 (2005)

[10] P.G. Burke, P. Francken and C. J. Joachain, J. Phys. B: **24,** 761 (1991)