Coherent control of atomic excitation using off-resonant strong few-cycle pulses.

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We study the dynamics of a two-level system driven by an off-resonant few-cycle pulse which has a phase jump ϕ at $t = t_0$, in contrast to many cycle pulses, under non rotating-wave approximation (NRWA). We give a closed form analytical solution for the evolution of the probability amplitude $|C_a(t)|$ for the upper level. Using the appropriate pulse parameters like phase-jump ϕ, jump time t_0, pulse width τ, frequency ν and Rabi frequency Ω_0 the population transfer, after the pulse is gone, can be optimized and for the pulse considered here, enhancement of $10^6 - 10^8$ factor was obtained.

Modern pulsed lasers produce bursts of light that are both ultra-short and ultra-strong, exhibiting durations comparable to those of molecular vibrations, and electric fields rivaling those near an atomic nucleus [1]. Attosecond lasers, emitting pulses with only a few optical cycles per pulse [2], hold the promise of controlling the phase difference between the carrier wave and its envelope[3].

The interaction between strong, broadband electromagnetic fields and atoms, especially laser radiation that is tuned far from resonance, is of current interest. Short pulses can excite coherence on high-frequency transitions that may be used for efficient generation of XUV radiation [4–6]. Shaped pulses can enhance transient population of the excited state [7] or create optimal coherence in TLS [8]. Recently we have found a new analytical solution describing the dynamics of a two-level atom under the action of laser radiation with an arbitrary pulse shape and polarization [9]. Furthermore, we have studied two mechanisms of atomic excitation: multi-photon excitation, and breaking of adiabaticity [4], and we have shown [10] that the latter can be the more efficient.

Interaction of such ultrashort pulses with a two-level atom under rotating-wave approximation does not give us the complete picture since the variation of the atomic polarization and population within the optical cycle is not slow. Thus we should not neglect the contribution of the counter-rotating terms in the Hamiltonian while studying few cycle pulses interaction with atomic systems [11–18]. On the other hand if the fields are not too strong and the variation of the atomic polarization and population within the optical cycle is slow, RWA appears to be a good approximation.

In this Brief report we studied the interaction of few-cycle pulses, in contrast to many cycle pulses [19–21], with two-level system. These pulse have a phase jump ϕ at $t = t_0$. Thus they can be characterized by the parameters peak Rabi frequency Ω_0, pulse width τ, carrier frequency ν, phase jump ϕ and jump moment t_0 along with the pulse envelope (which we have considered gaussian for the numerical simulation). We present an analytical solution for this problem. Using the appropriate characterizing parameters, the population transfer, can

be optimized and for the pulse considered here, enhancement of $10^6 - 10^8$ factor was obtained [see Fig. 5(b)].

The equation of motion for the probability amplitudes for the states $|a\rangle$ and $|b\rangle$ of a two-level atom (TLA) interacting with a classical field is given as [22]

$$\dot{C}_a = i\frac{\wp\mathcal{E}(t)}{\hbar}\cos(\nu t)e^{i\omega t}C_b, \qquad (1a)$$

$$\dot{C}_b = i\frac{\wp^*\mathcal{E}(t)}{\hbar}\cos(\nu t)e^{-i\omega t}C_a, \qquad (1b)$$

where $\hbar\omega$ is the energy difference between two levels, $\wp$ is the atomic dipole moment; $E(t) = \mathcal{E}(t)\cos\nu t$. In the rotating-wave approximation (RWA) we replace $\cos(\nu t)e^{\pm i\omega t} \rightarrow e^{\pm i\Delta t}/2$ where $\Delta = \omega - \nu$ [23], is detuning from resonance. Introducing $\Omega(t) = \wp\mathcal{E}(t)/\hbar$, Eq.(1) reduces to

$$\dot{C}_a = i\frac{\Omega(t)}{2}e^{i\Delta t}C_b, \qquad (2a)$$

$$\dot{C}_b = i\frac{\Omega^*(t)}{2}e^{-i\Delta t}C_a, \qquad (2b)$$

which have an integral of motion $|C_a|^2 + |C_b|^2 = 1$. If we define a function $f(t) = C_a(t)/C_b(t)$, Eq.(2) yields the following Riccati Equation

$$\dot{f} + i\frac{\Omega^*(t)}{2}e^{-i\Delta t}f^2 - i\frac{\Omega(t)}{2}e^{i\Delta t} = 0. \qquad (3)$$

Then $|C_a(t| = |f(t)|/\sqrt{1 + |f(t)|^2}$. Alternatively, we can get a second order linear differential equation for $C_a(t)$, from Eq.(2)

$$\ddot{C}_a(t) - \left[i\Delta + \frac{\dot{\Omega}(t)}{\Omega(t)}\right]\dot{C}_a(t) + \frac{|\Omega(t)|^2}{4}C_a(t) = 0. \qquad (4)$$

In this paper we will work without RWA, hence the Riccati Eq.(3) takes the new form as

$$\dot{f} + i\Omega^*(t)\cos(\nu t)e^{-i\omega t}f^2 - i\Omega(t)\cos(\nu t)e^{i\omega t} = 0. \qquad (5)$$

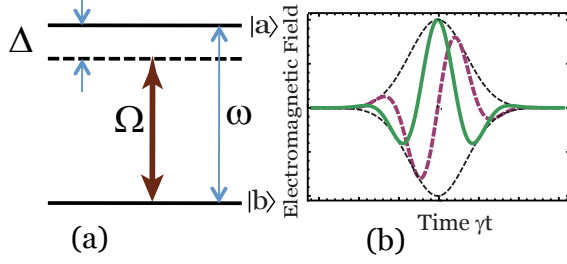The approximate solution for Eq.(5), in terms of the tip

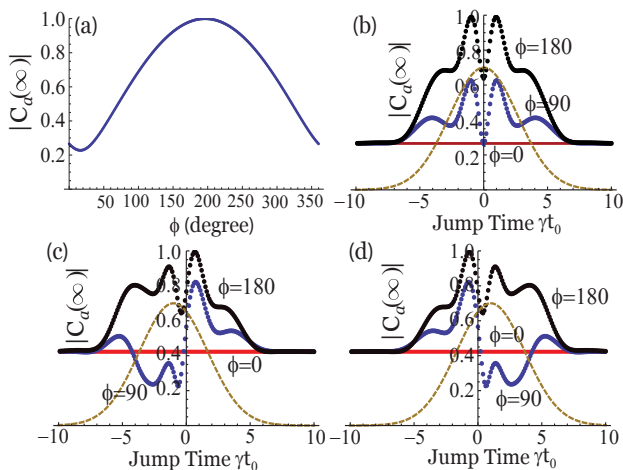FIG. 3: (Color Online) Effect of jump time t_0. (a) Here we have plotted the probability amplitude $|C_a(\infty)|$ against the phase jump ϕ. Phase jump is introduced at the peak of the gaussian envelope. (b) The symmetric influence on the degree of excitation with respect to the position of t_0. The symmetric response is lost for shifted gaussian input pulse (c) and (d). For numerical calculations we chose $\Omega_0 = 0.875\omega$, $\nu = 0.75\omega$, $\alpha = 0.331\omega$ and $\gamma = 1.25\omega$.

B. Effect of Pulse parameters: Numerical Analysis

In this section we will discuss the effect of the pulse parameters like phase jump time t_0, pulse witdth τ, detuning Δ and peak Rabi frequency Ω_0 on the degree of excitation of the upper level $|a\rangle$. For the computational purpose we have considered a Gaussian pulse of the form

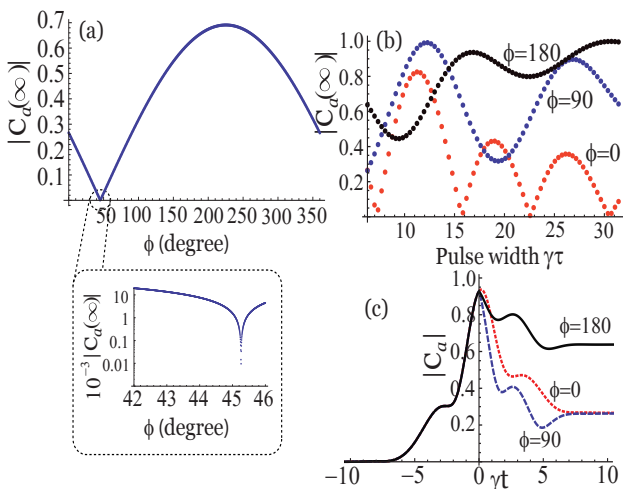FIG. 4: (Color Online) Effect of α. (a) Probability amplitude $|C_a(\infty)|$ varies in the range from $10^{-5} \sim 0.7$. (b) We have plotted $|C_a(\infty)|$ against normalized pulse width $\gamma\tau$ for fixed ω, ν, Ω_0 and three combinations of the phase jump $\phi = 0, \pi/2, \pi$. (c) Shows the temporal evolution for the three combinations used in (b). For numerical simulation we chose $\Omega_0 = 0.875\omega$, $\nu = 0.75\omega$, $\gamma = 1.25\omega$ and $\alpha = 0.331\omega$.
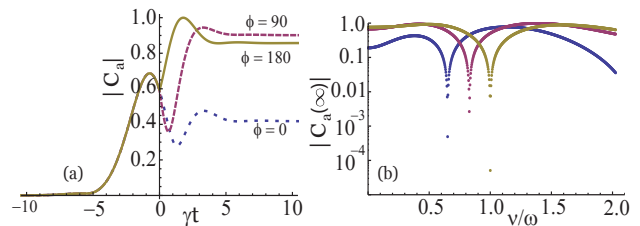

FIG. 5: (Color Online) (a) Temporal behavior of $|C_a|$ for difference combination of ϕ. (b) Plot of $|C_a(\infty)|$ against ν/ω. For numerical simulation we chose $\Omega_0 = 0.875\omega$, $t_0 = 0, \gamma = 1.25\omega$ and $\alpha = 0.331\omega$.

$\Omega(t) = \Omega_0 e^{-\alpha^2 t^2}$ where $\alpha = 2\sqrt{\ln 2}/\tau$ (τ is the FWHM of the pulse).

The main result showing the effect of relative position of t_0, with respect to the peak of the pulse, on the atomic excitation is shown in Fig. 3 and Fig. 7(a) where we have shown the dynamics of the two-level atom interacting with few-cycle pulse with a phase jump. In Fig. 3(a) we have one such scenario of $\phi = \pi/2$. Here the phase jump is introduced in the field at the peak of the gaussian envelope i.e $t_0 = 0$ and plotted the probability amplitude $|C_a(\infty)|$ against the phase jump ϕ. Interestingly the difference in the maximum and the minimum value corresponds to $\Delta\phi = \pi$. The symmetric nature of the atomic excitation is observed in Fig. 3(b) and the contour plot Fig. 7(a). With the shifted Gaussian pulse $\Omega = \Omega_0 e^{-\alpha^2(t \pm t_s)^2}$ [see Figs. 3(c) and 3(d)] the symmetry is lost. Also the effect of the phase jump becomes significant for t_0 within the FWHM of the pulse and gradually decreases when t_0 is close to the tail of the pulse. Identical response of the system, for $\gamma t_0 \approx 10$, is observed for three combinations of the phase jump $\phi = 0, \pi/2, \pi$.

While investigating the effect of few-cycle pulses on atomic systems, the parameter α plays an important role for a given value of the carrier frequency ν. It determines the number of cycles of the field in the pulse. The main results showing the effect of α or the pulse width τ is given in Fig. 4 and contour plot Fig. 7(b). If we look at the inset of Fig. 4(a) we see that the probability am-
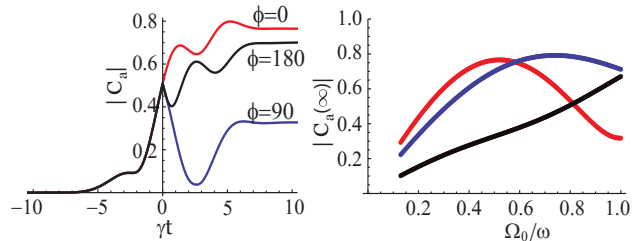

FIG. 6: (Color Online) (a) Temporal behavior of $|C_a|$ for difference combination of ϕ. (b) Plot of $|C_a(\infty)|$ against Ω_0. For numerical simulation in (b), we chose a shifted gaussian pulse with $t_s = 1$, $\nu = 0.75\omega$, $t_0 = 0, \gamma = 1.25\omega$ and $\alpha = 0.331\omega$. $\Omega_0 = 0.875\omega$ for Fig. (a).
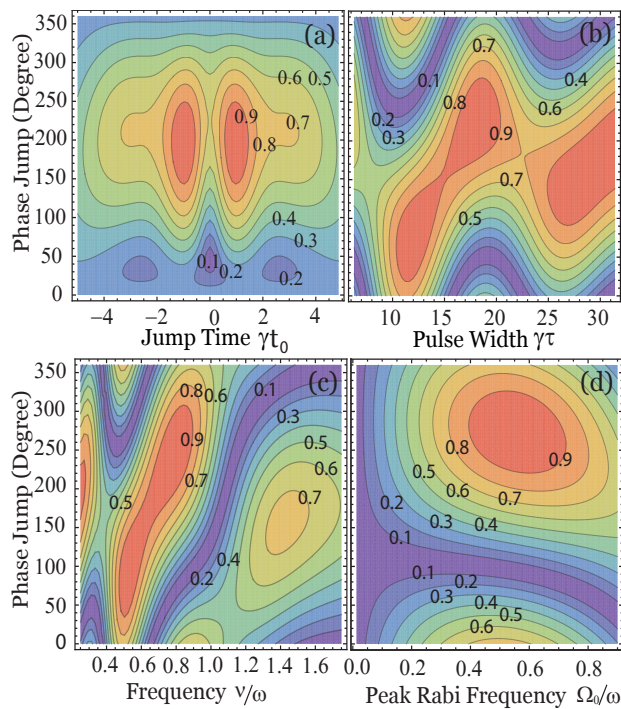
FIG. 7: (Color online) Contour plot showing the effect of pulse parameters like t_0, τ, ν and Ω_0 on the population left in the excited states $|a\rangle$ in (a), (b), (c) and (d) respectively. The influence of the phase jump time t_0 is symmetric with respect to t_0 is shown in (a). Parameters used are $\Omega_0 = 0.875\omega$, $\nu = 0.75\omega$, $\gamma = 1.25\omega,, t_0 = 0$ and $\alpha = 0.331\omega$ as required appropriately. For (c) we used $\alpha = 0.110\omega$

plitude $|C_a(\infty)|$ varies in the range from $10^{-5} \backsim 0.7$. In Fig 4(b) we have used three combination of phase jump ϕ ($\phi = 0, \pi/2, \pi$) to study the effect of α on the degree of excitation. For lower pulse width $(2 \leq \gamma\tau \leq 15)\phi = \pi/2$ creates more excitation than $\phi = 0$ or π.

In order to study the effect of detuning Δ we have plotted the response of the system in terms of $|C_a(\infty)|$ for the three combination of ϕ. Fig 5(a) shows the temporal behavior while Fig. 5(b) gives the information about steady-state population. The probability amplitude $|C_a(\infty)|$ varies in the range from $4.4 \times 10^{-4} \backsim 0.4$ for $\phi = 0$ and $5 \times 10^{-5} \backsim 0.9$ for $\phi = \pi$. When $|C_a(\infty)|$ is $\backsim 4.4 \times 10^{-4}$ for $\phi = 0$ we have $|C_a(\infty)| \backsim 1$ for $\phi = \pi$, thus we have an enhancement of $10^6 - 10^8$ factor in the population transfer by introducing a phase jump of π at the peak of the envelope function.

The effect of the peak Rabi frequency Ω_0 on the degree of excitation of the upper level in shown in Fig 6 and the contour plot Fig 7(d). While Fig 6(a) shows the temporal behavior of $|C_a|$ on the other hand Fig 6(b) gives the information about the population left in the upper level after the pulse is gone. We see that for some choice of Ω_0 $\phi = 0$ has the maximum effect while for some $\phi = \pi/2$ is dominant.

In conclusion, we have studied few-cycle pulses, with a phase jump ϕ at $t = t_0$, interacting with a two-level atom. This interaction is investigated without the rotating-wave approximation and we present an approximate solution for the probability amplitude $C_a(t)$ of the upper level. The approximate solution not only works well with multicycle pulse [9] but it is also in excellent agreement for few-cycles pulses [see Fig 2]. Using the appropriate pulse parameters ϕ, t_0, α and Ω_0 the population transfer, after the pulse is gone, can be optimized and for the pulse considered here, enhancement of $10^6 - 10^8$ factor was obtained [see Fig. 5(b)]

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