Ultra-short, off-resonant, strong excitation of two-level systems

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We present a model describing the use of ultra-short strong pulses to populate the excited level of a two-level quantum system. In particular, we study an off-resonance excitation with a few cycles pulse which presents a smooth phase jump i.e. a change of the pulse’s phase which is not step-like, but happens over a finite time interval. A numerical solution is given for the time-dependent probability amplitude of the excited level. The enhancement of the excited level’s population is optimized with respect to the shape of the phase transient, and to other parameters of the excitation pulse.

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I. INTRODUCTION

Ultra-strong pulses with intensities of the order of 1023 W/cm2, and duration of the order of attoseconds, with just few optical cycles, are feasible with present day technology (see e.g. [1–3]). This technological development has been motivated by the large number of possible applications, several of which rely on coherent population transfer techniques. A partial list of such applications is: stimulated Raman adiabatic passage (STI-RAP) [6–9], adiabatic rapid passage (ARP) [10], Raman chirped adiabatic passage (RCAP) [11, 12], temporal coherent control (TCC) [13, 14], coherent population trapping [15, 16], optical control of chemical reactions [17, 18], electronic population transfer techniques. A partial list of such applica-
tions is: stimulated Raman umklapp scattering [27], breakdown of dipole blockade obtained driving atoms by phase-jump pulses [28]. Moreover, recently two schemes for efficient and fast coherent population transfer have been presented [29], which use chirped and non-chirped few-cycles laser pulses. Another recent application [30] presents high-order harmonic generation obtained with laser pulses with a π-phase jump. Finally, the field of quantum information processing benefits from these results, since many qubit realizations rely on precise quantum levels manipulation [31–34].

The presence of few optical cycles in the pulse gives a constant phase difference between the carrier wave and the pulse shaped envelope [35], in contrast with many cycle pulses [28, 36, 37]. Moreover, optimizing the pulses parameters is proven to enhance the excited state population [38] or optimizing coherence in two-level systems (TLSs) [39]. In previous works we have already presented an analytical solution for the dynamics of a TLS excited with pulses of arbitrary shape and polarization [40, 41]. Since in the model we present the change rate of levels’ populations within a single optical cycle is not negligible, the rotating-wave approximation can’t be used. In other words in the present model we can’t neglect the contribution of the counter-rotating terms in the Hamiltonian [40, 41].

In a previous work [42] we have presented a similar model, representing the interaction of a TLS with few-cycle pulses, where at time \( t = t_0 \) the phase of the carrier wave jumps of an amount \( \phi \), this jump being sharp and step-like. In that work, numerical analysis of the analytic model has lead to an enhancement by a factor of \( 10^6 \) in the population transfer, with the optimal phase jump of \( \phi = \pi \) and the optimal time coincident with the peak of the envelope. In the present work we improve that model, considering a smooth phase change, i.e. not step-like but happening over a finite interval of time. This new model more closely describes a realistic experimental scenario.

The pulse is characterized by: Rabi frequency \( \Omega_\phi \), pulse width \( \tau \), carrier frequency \( \nu \), phase jump amplitude \( \phi \), phase jump time \( t_0 \) and phase jump duration \( \Delta t \). Moreover, we consider two qualitative parameters: the phase jump shape, and the pulse envelope shape.

![Image of three functional shapes of the smooth phase jumps](image)

**FIG. 1:** The three functional shapes of the smooth phase jumps used: the red is a dropping hyperbolic tangent: \( \phi(t) = (\pi/2)|1 - \tanh(5\alpha t)| \), the blue is a rising hyperbolic tangent: \( \phi(t) = (\pi/2)|1 + \tanh(5\alpha t)| \), and the black one is a hyperbolic secant: \( \phi(t) = (\pi/2) \text{sech}(\alpha t) \). In all the simulations the numerical normalized value is \( \alpha = 0.265 \).

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We present an analytical solution for the time evolution of the excited state’s population, together with a numerical simulation. In the numerical simulation we use 3 functional shapes for the smooth phase jump: rising hyperbolic tangent, dropping hyperbolic tangent, and gaussian peak, (see figure 4), whereas for the envelope a gaussian peak has been used. Numerically optimizing the pulses parameters we have obtained enhancements for the population transfer of the order of $10^4$.

A. NUMERICAL SIMULATION AND DISCUSSION

Be $|a\rangle$ and $|b\rangle$ the states of a two-level atom (TLA), with energy difference $\hbar\omega$, and atomic dipole moment $\varphi$. If we let this system interact with a classic field $E(t) = E(t)\cos(\nu t)$, the equations of motion for the relative wavefunctions are [43]:

$$\dot{C}_a = i\frac{\varphi E(t)}{\hbar}\cos(\nu t) e^{i\omega t} C_b,$$

$$\dot{C}_b = i\frac{\varphi E(t)}{\hbar}\cos(\nu t) e^{-i\omega t} C_a,$$

where $\Delta = \omega - \nu$ is the detuning from resonance. Similarly to [42], defining $f(t) = C_a(t)/C_b(t)$ and $\Omega(t) = \varphi E(t)/\hbar$, we have the following Riccati equation:

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

(2)

The approximate solution for Eq. (2), in terms of the tip angle $\theta(t)$ is given as in [40]:

$$f(t) = i \int_{-\infty}^{t} dt' \left\{ \frac{d\theta(t')}{dt'} - \theta^2(t') \frac{d\theta'(t')}{dt'} \right\} \times \exp \left[ 2 \int_{t'}^{t} \theta(t'') \theta'(t'') dt'' \right],$$  

(3)

where the tip angle $\theta(t)$ has been defined as

$$\theta(t) = \int_{-\infty}^{t} \Omega(t')\cos(\nu t') e^{i\omega t'} dt'$$  

(4)

from which we have $|C_a(t)| = |f(t)| / \sqrt{1 + |f(t)|^2}$. What is of interest is the asymptotic behavior of $|C_a(\infty)|$. In [42] it is shown good agreement between the analytical and a numerical simulation. To introduce the phase jump, we can write the Rabi frequency as

$$\Omega(t) = \Omega_0(t) \cos \nu t e^{i\omega t_0} e^{i\phi(t)}$$  

(5)

and then, using the same method as in [42], we can obtain an approximated analytic solution for the Riccati equation (2):

$$f(t) = i \int_{-\infty}^{t} dt' \Phi(t') \exp \left[ 2 \int_{t'}^{t} \zeta(t'') dt'' \right],$$  

(6)

The approximate analytical solution is in good agreement with the numerical simulation obtained by directly solving the coupled differential Eq.(1). From Fig.(3) we see that even for complex phase function the agreement is good. For the sake of completeness, we have added an appendix in which we show the strength of this approach beyond standard TLS. Indeed the Riccati equation approach gives a closed compact from which

![Graph](image_url)
FIG. 4: In this figure we present the results of the numerical analysis. Each of the three rows of plots refers to a different functional shapes of the smooth phase jump (phase change function). For each row we have a plot of the smooth phase jump, a plot of the excited state’s population as a function of time, and a plot of the excited state’s population left after the pulse is gone as a function of the normalized excitation frequency. Similarly to figure [3], the functional shape of the excitation pulse is \( \Omega(t) = A e^{-\alpha^2 t^2} e^{i \theta(t)} \). Moreover, for the plots of the excited state’s population as function of time we have used the numerical value of \( \nu/\omega = 0.75 \). Phase change of the form (a) \( \phi(t) = (\pi/2)[1 + \tanh(\alpha_1 t)] \), with three different values of \( \alpha_1 \): red (steeper) \( \alpha_1 = 5\alpha \); blue (in-between) \( \alpha_1 = \alpha \); black (smoother) \( \alpha_1 = 0.5\alpha \). (b) Corresponding behavior of the excited level population \( |C_1(t)| \). (c) Asymptotic value of the excited state population, as a function of the “resonance ratio” (excitation’s frequency divided by transition’s frequency) for this form of the phase change. (d) Phase change of the form \( \phi(t) = (\pi/2)[1 - \tanh(\alpha_1 t)] \), with (as in (a)) three different values of \( \alpha_1 \): red (steeper) \( \alpha_1 = 5\alpha \); blue (in-between) \( \alpha_1 = \alpha \); black (smoother) \( \alpha_1 = 0.5\alpha \). (e) Corresponding behaviour of the excited level population \( |C_1(t)| \). (f) Asymptotic excited population for this form of the phase change. (g) Phase change of the form: \( \phi(t) = (\pi/2) \sech^2(\alpha_1 t) \), \( \alpha_1 \): red (larger) \( \alpha_1 = \alpha \); blue (in-between) \( \alpha_1 = 10\alpha \); black (narrower) \( \alpha_1 = 20\alpha \). (h) Corresponding behaviour of the excited level population \( |C_1(t)| \). (i) Asymptotic excited population for this form of the phase change. The value used for \( \alpha \) is \( \alpha = 0.265 \). For numerical simulations we chose \( A = 0.04375\omega \), \( \alpha = 0.265y \) and \( y = 1.25\omega \) where \( \omega = (2\pi) 80 \text{ GHz} \).

Both the temporal and steady-state behavior of the two and three-level system can obtained.

An interesting observation is that it is possible to rewrite the Rabi frequency in (5) as

\[
\Omega(t) = \Omega_0(t) \cos \nu t e^{i(\omega t + \phi(t))} \tag{7}
\]

and then define \( \tilde{\omega}(t) = \omega + \phi(t)/t \) and interpret this as a modulation of the atomic frequency, instead of a modulation of the excitation. Experimentally this can be realized in several ways, e.g. using modulated Zeeman or Stark effect.

Now we move to discuss our numerical simulation of the dynamics of the two-level atom interacting with ultra-short, off-resonant and gradually changing phase.
\(\phi(t)\). We have performed numerical solution of the Riccati equation, using different types of phase change (smooth phase jump) functions. The result of this numerical analysis is shown in figure 3. The goal of this study is to find the best phase change which allows for the best coupling (most efficient energy exchange) of the excitation pulse with the excited state.

In figure 4 we present the results of the numerical analysis. Each of the three rows of plots refers to a different functional shapes of the smooth phase jump (phase change function). For each row we have a plot of the smooth phase jump, a plot of the excited state’s population as function of time, and a plot of the excited state’s population left after the application of the pulse as a function of the normalized excitation frequency. Similarly to figure 3 the functional form of the excitation pulse is \(\Omega_0(t) = A e^{-at^2} \phi(t)\). Moreover, for the plots of the excited state’s population as function of time we have used the numerical value of \(\nu/\omega = 0.75\).

For this numerical simulation we have considered the following three phase functions (a)(b)(c): \(\phi(t) = (\pi/2) |1 + \tanh(at)|\), (d)(e)(f): \(\phi(t) = (\pi/2) |1 - \tanh(at)|\) and (g)(h)(i): \(\phi(t) = (\pi/2) \sech^2(at)\). We can see how the phase change duration \(\Delta t\), i.e. the steepness of the \(\phi(t)\) function, has not an unique effect on the excited population, and depends on the general shape of the phase change. In particular, it is worth noting that for ascending and descending phase changes built on the \(\tanh(t)\) function the effect of the steepness is opposite. We can observe a global behaviour which relates the characterizing parameters of the phase change with the amplitude of the population of the excited state. Qualitatively, for the ascendent hyperbolic tangent we observe that by increasing the slope the population increases. On the other hand, for the descendent hyperbolic tangent the effect of this parameter is reversed: decreasing the slope of phase change leads to a decrease of the population. We remark that these behaviors are only global, and are reversed for some small ranges of frequencies. As an example, in plot 4.(f), for low ranges of laser frequencies, by decreasing the slope we increase the population, which is opposite of the behavior observed for higher frequencies. For the peaked shape (sech(t)), no general behaviors are observed. However, for the intermediate range of frequencies it can be observed a link between the increasing of the pulse width and the increase of the population.

\[C(t) = \frac{i}{\Omega_0(t)} \frac{dC(t)}{dt} + \frac{1}{2} \left[ C(t) \frac{dC(t)}{dt} - \frac{1}{2} \right] \]  
(A1)
In terms of $f$ and $g$ Eqs. (A1, A2, A3) reduces to
\[ f'(t) + i\tilde{\Omega}_1 f^2(t) = \tilde{\Omega}_1 + i\tilde{\Omega}_2 g(t) \] (A6)
\[ g'(t) + i\tilde{\Omega}_1 f(t)g(t) = i\tilde{\Omega}_2 f(t) \] (A7)

In order to solve these equations we extended the method developed [40, 41]. By neglecting the non-linear term $f^2(t)$ and the term $\propto g(t)$ in Eq. (A6) we can solve for $f_1(t)$ as
\[ f_1(t) = i \int_{-\infty}^{t} \tilde{\Omega}_1 dt' \] (A8)

Similarly by neglecting the term $\propto g(t)$ in Eq. (A7) we can solve for $g_1(t)$ as
\[ g_1(t) = -\int_{-\infty}^{t} \tilde{\Omega}_2 (t')\theta_1(t')dt' \] (A9)

where the tip angle $\theta_1(t)$ is defined as
\[ \theta_1(t') = \int_{-\infty}^{t'} \tilde{\Omega}_1 dt' \] (A10)

Next let us write the non-linear term in Eq. (A6) as
\[ f^2(t) = [f(t) - f_1(t)]^2 + 2f(t)f_1(t) - f_1^2(t) \] (A11)

Then Eq. (A6) can be written as
\[ f(t) + i\tilde{\Omega}_1 f(t)[f(t) - f_1(t)]^2 + 2f(t)f_1(t) - f_1^2(t) = i\tilde{\Omega}_1 + i\tilde{\Omega}_2 g(t) \] (A12)

Let us assume that $g(t) \approx g_1(t)$ and we neglect $[f(t) - f_1(t)]^2$ in this case we can write Eq. (A12) in term of the tip angles $\theta_1(t)$ and $\theta_2(t)$
\[ f(t) + i\tilde{\Omega}_1 f(t)\{2f(t)f_1(t) - f_1^2(t)\} = i\tilde{\Omega}_1 + i\tilde{\Omega}_2 g_1(t) \] (A13)

where
\[ \theta_2(t') = \int_{-\infty}^{t} \tilde{\Omega}_2 (t')dt' \] (A14)

The analytical solution of the equation Eq. (A13) is then:
\[ f(t) = e^{-a(t)}\int_{t_0}^{t} b(t')e^{a(t')}dt' \] (A15)

where
\[ a(x) = 2i\theta_1(t)f_1(t) \] (A16)

and
\[ b(x) = i\theta_1(t) + i\theta_2(t)g_1(t) + i\theta_1(t)f_1^2(t) \] (A17)

For $g(t)$ the solution can be obtain from Eq. (A7) where we use $f(t) = f_1(t)$:
\[ g(t) + i\tilde{\Omega}_1 f_1(t)g(t) = i\tilde{\Omega}_2 f_1(t) \] (A18)

which give us
\[ g(t) = e^{-a(t)}\int_{t_0}^{t} D(t')e^{a(t')}dt' \] (A19)

where
\[ c(x) = i\tilde{\Omega}_1 f_1(t) \] (A20)

and
\[ D(x) = i\tilde{\Omega}_2 f_1(t) \] (A21)

In Fig. 5 we have plotted the Numerical (red dotted line) and analytical (blue solid line) solutions of the amplitude of the state $|\psi\rangle$ after long time in function of $\nu/\omega$ for the laser pulse envelopes $\Omega_1(t) = \Omega_2(t) = \Omega_0 \text{sech}(at)$. For numerical simulation we chose $\Omega_0 = .04\omega, \alpha = 0.075\omega, \omega_{\text{db}} = \omega_{\text{dc}} = \omega = 1$. We see that the approximate analytical solution matches well with the numerics under the parameters considered here. Extension of this methodology to Schrodinger equation [52, 53] and position dependent mass Schrodinger(PDMSE) equation can be found in [54, 55].
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