Energy efficient method for two-photon population transfer with near-resonant chirped pulses

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Abstract: We investigate a method for complete population inversion in three level systems through π-pulse bichromatic two-photon coherent excitation and study the dependence on the chirp of the laser pulses. We observe that the population inversion does not monotonously decrease with increasing the time-bandwidth product, and that the excitation depends on the sign of the chirp of the individual pulses. Our results evidence a strategy for coherent population transfer which is energetically superior to adiabatic methods and opens the door for real-world applications, since it alleviates the need for challenging generation of transform-limited pulses at arbitrary wavelengths.

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1. Introduction

Steering quantum processes in atoms and molecules through the manipulation of the properties of optical fields is the goal of coherent quantum control [1–6]. In particular, control of population transfer in multiphoton transitions with laser pulses has been achieved through several methods. A common strategy for ultrashort pulses is the manipulation of the spectral phases and amplitudes of the frequency components of the fields exciting the medium, resulting in pulses or sequences of pulses with very diverse temporal shapes which enhance or suppress the coupling between selected states by exploiting quantum interference effects [7–14]. Other techniques using longer pulses such as adiabatic methods [15–18] and π-pulse polychromatic control [19–23] can be implemented to induce complete population transfer between a pair of quantum states. Multiphoton control techniques are valuable in several areas, including nonlinear spectroscopy [24], in femtochemistry and biology [25], or in quantum information and in the quantum engineering of light states [26], where quantum systems need to be fully controlled, among others.

In recent work [21–23] it was shown theoretically and experimentally that complete population inversion in atomic sodium can be realized by conveniently choosing the energy and

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duration of pulses near-resonant with the dipole allowed transitions in a three-level configuration (see Fig. 1). Two efficient schemes for two-photon coherent excitation were presented; one of them considered bichromatic pulse pairs separated by a particular delay time, and the other considered excitation by single bichromatic pulses. Relevant in these two-photon excitation schemes is the fact that the intensities required for optimal excitation are on the order of the intensities for resonantly pumped single-photon transitions, and therefore they result energetically more efficient than adiabatic methods, as it will be further explained below. In the present paper we investigate the influence of pulse chirp, as it occurs in most real-world situations, on complete population inversion on the example of a three-level system. Such study is important since the necessity to generate pairs of transform-limited pulses at various wavelengths prevented widespread application of multi-color π-pulse excitation even though it is energetically superior to most other methods. We show that the population transfer does not monotonously decrease with increasing frequency chirp, and that the occupation probability of the highest state depends on the sign of the chirp of the individual pulses.

2. Theory, results and discussion

The experimental results [21–23] were compared to both a simple three-level and a complete 32-level system including the hyperfine structure of the involved atomic sodium transitions. When the detuning from the intermediate level is chosen sufficiently large compared to the width of the sodium hyperfine structure, the three-level approximation is in good agreement with the more complex treatment. The coherent interaction between a three-level system and two fields has been analyzed for certain processes [27–34]. For instance, in the particular case of each field being in resonance with the intermediate state, the pulse area of each pulse for complete transfer of the occupation probability from the ground state to the highest state is simply $\sqrt{2}\pi$, with the pulse area defined as $\int_{-\infty}^{\infty}\Omega(t)dt$, where $\Omega(t) = \mu E(t)/\hbar$ is the Rabi frequency of the transition, μ is the electric dipole moment and $E(t)$ is the electric field amplitude of the laser pulse. When considering different shaped fields and detunings, however, the system needs to be integrated numerically.

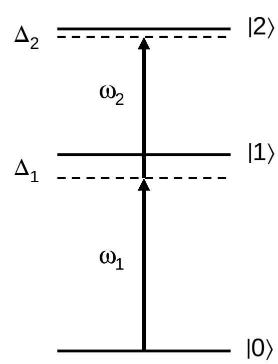

Fig. 1. Schematic energy level configuration.

In this work we consider a three-level system in a cascade-configuration (Fig. 1) coherently driven by near-resonant single bichromatic laser pulses. In order to investigate the transfer of population with arbitrarily chirped and detuned pulses we numerically integrate the density matrix equations beyond the rotating-wave approximation. The model reads as follows

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$$
\begin{aligned}
\dot{\rho}_{00} &= \frac{iE(t)}{\hbar}\{d_{01}(\rho_{10}-\rho_{01})\}, &(1)\\
\dot{\rho}_{11} &= \frac{iE(t)}{\hbar}\{d_{01}(\rho_{01}-\rho_{10})+d_{12}(\rho_{21}-\rho_{12})\}, &(2)\\
\dot{\rho}_{22} &= \frac{iE(t)}{\hbar}\{d_{12}(\rho_{12}-\rho_{21})\}, &(3)\\
\dot{\rho}_{01} &= i\omega_{10}\rho_{01}+\frac{iE(t)}{\hbar}\{d_{01}(\rho_{11}-\rho_{00})-d_{12}\rho_{02}\}, &(4)\\
\dot{\rho}_{02} &= i\omega_{20}\rho_{02}+\frac{iE(t)}{\hbar}\{d_{01}\rho_{12}-d_{12}\rho_{01}\}, &(5)\\
\dot{\rho}_{12} &= i\omega_{21}\rho_{12}+\frac{iE(t)}{\hbar}\{d_{12}(\rho_{22}-\rho_{11})-d_{01}\rho_{02}\}. &(6)
\end{aligned}
$$

The interaction time in all the cases of our study is shorter than any decay time of the system, and therefore all decays of the levels and coherence dephasings have been neglected. In Eqs. (1)-(6), ρ_{00}, ρ_{11} and ρ_{22} are the populations of levels 0, 1 and 2, respectively, ρ_{01}, ρ_{02} and ρ_{12} are the coherences between the energy levels, d_{01} and d_{12} are the dipole coupling coefficients of the dipole allowed transitions, and $\omega_{ij} = |E_j - E_i|/\hbar$ are the angular frequencies of the transitions, with E_i being the energies of the different quantum states. The laser pulses are of Gaussian shape, with the electric field $E(t)$ given by

$$
E(t) = e^{-(t/\tau)^2}\{E_1\cos[\omega_1 t - a_1(t/\tau)^2] + E_2\cos[\omega_2 t - a_2(t/\tau)^2]\}. \qquad (7)
$$

We consider both frequency components of the bichromatic pulse having the same duration, with $\tau = t_p/(\sqrt{2\ln 2})$, where t_p is the full width at half maximum of the pulse intensity profile. E_{01} and E_{02} are the peak amplitudes of the pulse components, ω_1 and ω_2 are the optical angular frequencies, and a_1 and a_2 are the respective adimensional chirp parameters. Note that the chirp parameters have been chosen such that for $a_i > 0$ the pulses are downchirped, i.e., with $a_1 > 0$ and $a_2 > 0$ we have down-down chirped pulses, with $a_1 < 0$ and $a_2 > 0$ we have up-down chirped pulses, etc, with the chirp values in s^{-2} being a_i/τ^2. In Fig. 1, $\Delta_1 = \omega_{01} - \omega_1$ and $\Delta_2 = \omega_{02} - (\omega_1 + \omega_2)$ are the detunings associated to the transitions.

The level structure that we have chosen corresponds to the energy levels of the atomic sodium $^{23}_{11}Na$, in particular our ground state $|0\rangle$ represents the sodium $3s_{1/2}$ state, our intermediate state $|1\rangle$ is the $3p_{3/2}$ state, and the upper state $|2\rangle$ is the $4s_{1/2}$. We consider values for the field detunings that are larger than the width of the hyperfine structure of the energy levels in sodium, and hence we can neglect the complex energy level structure. The goal being complete population transfer from the $3s_{1/2}$ to the $4s_{1/2}$ states, starting from the ground state, we consider a bichromatic pulse with a duration of 10 ps, composed of two components with approximate central wavelengths of 589 nm and 1140 nm. The detunings are $\Delta_1 = 94.14$ GHz and $\Delta_2 = 10.95$ GHz. When the pulses are transform-limited ($a_1 = a_2 = 0$), complete population inversion is obtained when the peak amplitudes are $E_1 = 3.25 \times 10^6$ V/m and $E_2 = 4.34 \times 10^6$ V/m. The dipole coupling coefficients are $d_{01} = d_{12} = 1.85 \times 10^{-29}$ Cm. In this situation, the population in state $|2\rangle$, after the medium has interacted with the bichromatic pulse, is higher than 99.7%. The pulse that achieves total population transfer from the ground state $|0\rangle$ to the upper state $|2\rangle$ is called *bichromatic π-pulse*, in analogy with regular π-pulses in resonant two-level systems [35].

As commented above the excitation by π-pulses is energetically superior than most adiabatic methods. To illustrate this fact we reduce our system to an pseudo-two-level system, which a good approximation when the intermediate level does not get populated in the process and

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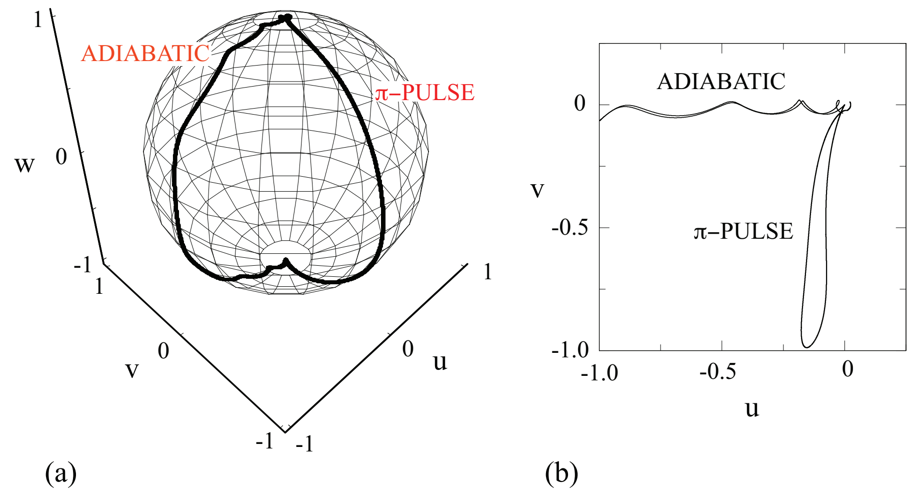

Fig. 2. Evolution of the normalized amplitude of the Bloch vector (u,v,w) on the sphere for π-pulse and adiabatic excitation, as indicated, for the three-level system reduced to a pseudo-two-level system (a). Projection of the two trajectories onto the u, v plane (b).

therefore when the detuning Δ_1 is sufficiently big to treat the system with first order perturbation theory. In the rotating-wave approximation and by adiabatically eliminating the intermediate level from Eqs. (1)-(6), the resulting two-level system can be written as follows

$$\dot{\mathrm{u}} = -\frac{\Omega_1^2-\Omega_2^2}{\Delta_1}\mathrm{v}, \tag{8}$$

$$\dot{\mathrm{v}} = \frac{\Omega_1^2-\Omega_2^2}{\Delta_1}\mathrm{u}+\frac{2\Omega_1\Omega_2}{\Delta_1}\mathrm{w} \tag{9}$$

$$\dot{\mathrm{w}} = -\frac{2\Omega_1\Omega_2}{\Delta_1}\mathrm{v}, \tag{10}$$

where u is the real part of the two-photon coherence ρ_{02} accounting for dispersive processes in the pseudo-two-level system, v is its imaginary part and is related to absorption, and w is the population inversion $\rho_{22}-\rho_{00}$. Ω_1 and Ω_2 are the Rabi frequencies of the transitions $0-1$ and $1-2$, respectively. In the derivation of Eqs. (8)-(10) we have considered $\Delta_2=0$ for simplicity. Figure 2 shows the evolution of the normalized amplitude of the Bloch vector (u,v,w) [35] obtained by numerical integration of the three-level system (1)-(6). The two trajectories plotted on the sphere, as indicated, correspond to optimal bichromatic π-pulse excitation of the system with the parameters defined above, and to adiabatic excitation with the same material and detuning parameter values but using suited field amplitudes ($E_1=E_2=1.7\times10^7$ V/m) and pulse delay (10 ps) to obtain optimal adiabatic population transfer [17]. As shown in the Bloch sphere representation (Fig. 2), π-pulse excitation is basically absorptive, contrarily to

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the adiabatic process which is mainly dispersive. Therefore, since the absorptive component v remains small during the interaction, adiabatic processes need more laser power to induce a comparable population inversion, as it is apparent from Eq. (10).

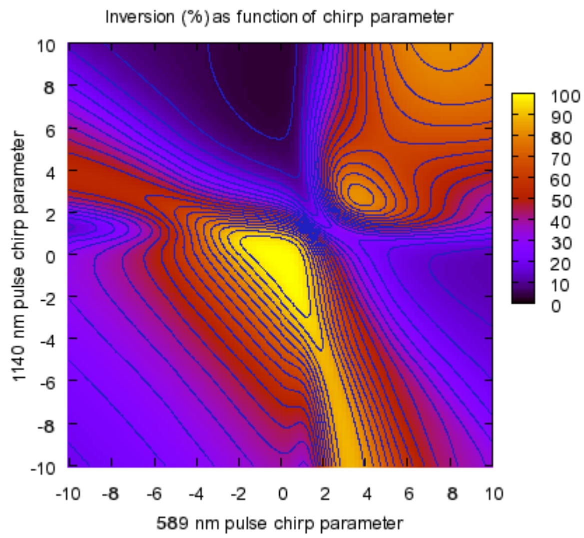

Fig. 3. Final value of the population ρ_{22} (in %) as a function of the adimensional chirp parameter for the 589 nm (a_1) and the 1140 nm (a_2) bichromatic pulse components. For instance, $a_i = 10$ gives a chirp value of ≈ 0.14 ps^{-2}. Other parameters are $t_p = 10$ ps, $E_1 = 3.25 \times 10^6$ V/m, $E_2 = 4.34 \times 10^6$ V/m, $\Delta_1 = 94.14$ GHz and $\Delta_2 = 10.95$ GHz.

The main results of our numerical integration, once we take chirp of the pulses into account, are summarized in the contour map in Fig. 3. Shown is the final value of the population in state $|2\rangle$ (ρ_{22}) as function of the chirp parameter of the pulses; we keep the pulse area constant while changing the chirp parameter. It can be seen that, contrarily to what would be naively expected, the final population in level $|2\rangle$ does not monotonously decrease with increasing the time-bandwidth product of the pulses. Instead, we observe a reach behavior which depends not only on the time-bandwidth product of the pulses but also on the sign of the chirp. When both are positive ($a_1 > 0$ and $a_2 > 0$), we observe a modulation of the final value of ρ_{22} as the chirp increases with $a_1 = a_2$. This can also be seen in Fig. 4, where we have plotted the final value of the populations ρ_{00}, ρ_{11} and ρ_{22} as a function of the chirp parameter for $a_1 = a_2$. The black full line in Fig. 4 representing ρ_{22} is therefore a diagonal cut of the contour map of Fig. 3. Clearly, the behavior for downchirped pulses ($a_1 > 0$ and $a_2 > 0$) is different from the behavior obtained when considering upchirped pulses ($a_1 < 0$ and $a_2 < 0$); in this latter case, the final population in level $|2\rangle$ does decrease monotonously as the time-bandwidth product increases (see the behavior for negative values of a_1 and a_2 in Figs. 3 and 4). Note also the results for pulses having different chirp sign: We observe a nice lightened band when the value of a_2 is increased with negative sign and the value of a_1 stays between 2 and 4 (see the right-bottom area in Fig. 3). We finally note that in all cases, for each value of a_1 and a_2, the amplitudes of the fields could be lightly tuned to give a somewhat higher population in level $|2\rangle$.

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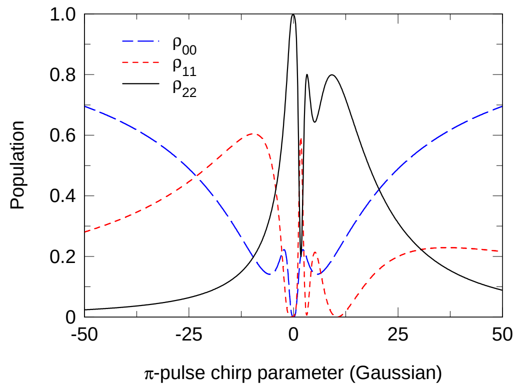

Fig. 4. Final value of the populations ρ_{00}, ρ_{11} and ρ_{22} (as indicated) as a function of the chirp parameter (in units of τ^{-2} i.e. $2\ln 2/100ps^2$) for $a_1 = a_2$. Other parameters are as in Fig. 3.

3. Conclusion

We have investigated the influence of frequency chirp on the complete population transfer produced by a bichromatic π-pulse with a detailed study on the example of a three-level system.

Our study gives valuable ranges of parameters for real-world applications thereby alleviating the need for complex generation of various pulse pairs at different wavelengths with given time-bandwidth product, such was the scope of previous work [21–23]. We have confirmed that frequency chirp in the bichromatic pulses can be used to achieve complete population inversion in a two-photon coherently driven three-level system. We have shown that the final population in the highest level of the three-level system due to the interaction with the near-resonant pulses does not monotonously decrease with increasing the time-bandwidth product when both pulses are equally downchirped, although it does decrease monotonously when they are both upchirped. We have found regions in the space of the chirp parameters in which the transfer of the occupation probability from the ground state to the highest state is fairly good although the pulses are far from being transform-limited. We have also illustrated, by using a Bloch sphere representation, that π-pulse excitation is energetically more efficient than most adiabatic processes. Our results might be extended to the case of more complex multilevel systems, atoms or molecules, and they can be of value for coherent quantum control schemes and in areas where energy quantum states need to be individually addressed and populated.

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