Ultrafast population transfer in three-level Λ systems driven by few-cycle laser pulses

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(Received 16 July 2001; published 14 November 2001)

The maximal population transfer in three-level Λ system driven by linear polarized few-cycle laser pulses is investigated numerically. A time-dependent Schrödinger equation without the rotating wave approximation is solved. The maximal population transfer depends critically on the chirp rate and the intensity of the pulses. Almost complete population transfer can be achieved if the difference between the transition dipole moments is not very large.

DOI: 10.1103/PhysRevA.64.065402 PACS number(s): 42.50.Hz, 42.65.Re

Recent technological advances in ultrafast optics permit to generate pulses shorter than 10 fs conveniently [1]. These pulses contain only a few optical cycles of the carrier frequency and their spectrum width can reach several hundred nanometer. The few-cycle pulses present novel phenomena in the field of light-matter interaction. Several important applications include high-order harmonic and attosecond x-ray pulse generation [1]. In this paper, we will study the population transfer in the three-level atom interacted with the few-cycle pulses laser based on numerical simulation.

The coherent population transfer in three-level atoms has been extensively studied in recent years [2–14]. This phenomenon contains most of the basic properties of multilevel coherence and has a great potential for realistic application. As an example, stimulated Raman adiabatic passage (STIRAP) is a very robust method to produce complete population transfer in the three-level system. The STIRAP technique uses the counterintuitively delayed laser pulses to adiabatically correlate the initial populated level to the desired target level during the system evolution. STIRAP technique has been implemented with continuous or pulsed lasers [2]. Generally, the adiabatic condition limits the pulse width in the nanosecond time scale. However, as pointed out in Ref. [8], the use of ultrashort pulse lasers to obtain population transfer may have some special benefits, such as, easy to realize the transition in the UV or vacuum ultraviolet (vuv) energy range, or control the level population distribution in the ultrafast process. For pulses in the picosecond or subpicosecond regime, adiabatic condition can be satisfied by chirping the frequency of the laser pulses and nearly complete population transfer has been produced [3,4,8,13,14]. Most of these methods are based on the rotating wave approximation (RWA) and the adiabatic condition is needed to satisfy. However, RWA is not valid for interaction of few-cycle pulses with atoms [15,16]. At present, it is not clear whether a complete population transfer is possible in the interaction of few-cycle pulses with three-level Λ atoms. This problem is investigated numerically in this paper.

The three-level Λ system under consideration is shown in Fig. 1. The 1-2 transition frequency is ω_1 and the 2-3 transition frequency is ω_2. The transition between level 1 and 3 is electric dipole forbidden. Only one laser field is interacted with the system. The full Hamiltonian is

$$H(t)=H_0+\vec{d}\cdot\vec{E}(t), \quad (1)$$

where the free Hamiltonian $H_0=\mathrm{Diag}(-\omega_1,0,-\omega_2)$, $\vec{d}$ is the dipole moment operator and $\vec{E}(t)$ is the electric field of the laser,

$$\vec{E}(t)=\vec{e}E_0 s(t)\cos(\omega t+1/2\chi t^2), \quad (2)$$

where $\vec{e}$ is the unit polarization vector, ω is the carrier frequency, χ is the chirp rate, and E_0 is the maximal electric-field amplitude. $s(t)$ is the shape function. For convenience, we assume it is the Gaussian function

$$s(t)=\exp(-t^2/\tau^2),$$

with τ the parameter of the pulse width.

To characterize the Hamiltonian matrix, we introduce the Rabi frequency $\hbar\Omega=-\langle 1|\vec{d}\cdot\vec{e}|2\rangle E_0$ and define the ratio of the transition dipole moments $\gamma=\langle 2|\vec{d}\cdot\vec{e}|3\rangle/\langle 1|\vec{d}\cdot\vec{e}|2\rangle$. The Hamiltonian now has the formula

$$H(t)=\begin{bmatrix} -\omega_1 & \Omega s(t)\cos(\omega t+1/2\chi t^2) & 0 \\ \Omega s(t)\cos(\omega t+1/2\chi t^2) & 0 & \gamma\Omega s(t)\cos(\omega t+1/2\chi t^2) \\ 0 & \gamma\Omega s(t)\cos(\omega t+1/2\chi t^2) & -\omega_2 \end{bmatrix}. \quad (3)$$

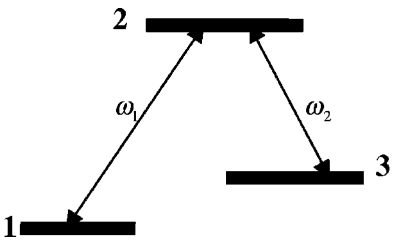

FIG. 1. Three-level Λ system. The transition between initial level $|1\rangle$ and the final level $|3\rangle$ is electric dipole forbidden.

The evolution of the probability amplitudes of the three levels is determined by the time-dependent Schr'odinger equation,

$$i\hbar\frac{d}{dt}\begin{bmatrix}c_1\\c_2\\c_3\end{bmatrix}=H(t)\begin{bmatrix}c_1\\c_2\\c_3\end{bmatrix}. \tag{4}$$

Suppose at the initial time the three-level system is in its ground level $|1\rangle$,

$$c_1(-\infty)=1,\quad c_2(-\infty)=0,\quad c_3(-\infty)=0,$$

we are interested in the population distribution at $t\rightarrow+\infty$, $P_n=|c_n(+\infty)|^2(n=1,2,3)$.

In Eqs. (3) and (4), we have neglected the spontaneous emission from the intermediate level $|2\rangle$ since the interaction time is only a few cycles of the carrier frequency. Eqs. (3) and (4) are exactly quantum and the RWA is not used. We will numerically solve Eq. (4) to investigate the problem of population transfer.

In our simulation, the carrier frequency of the laser pulse is $\omega=0.75\pi/\text{fs}$ corresponding to 800 nm central wavelength and $\tau=5$ fs so that the full-width-at-half-maximum pulse width is 5.9 fs. The frequency detunings are $\omega_1-\omega=0.05\pi/\text{fs}$ and $\omega_2-\omega=-0.05\pi/\text{fs}$. Both transition frequencies are contained in the bandwidth of the pulse.

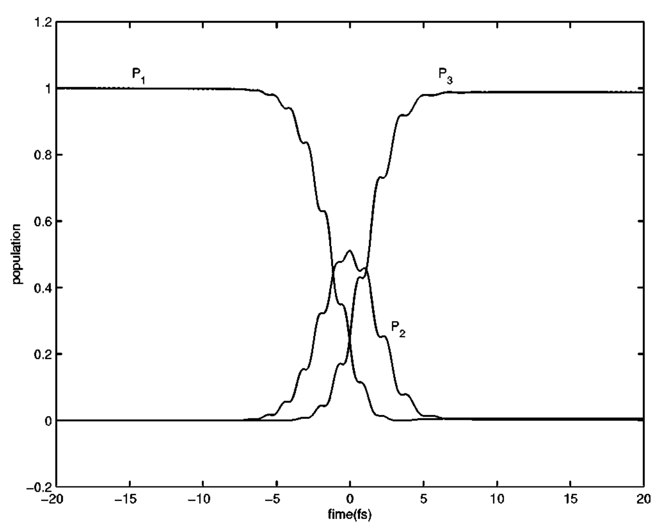

FIG. 2. Population evolution $P_n(t)$ for $n=1,2,3$, with $\Omega=0.56$ fs^{-1}, $\chi=0.04$ fs^{-2} for $\tau=5$ fs and $\gamma=1$.

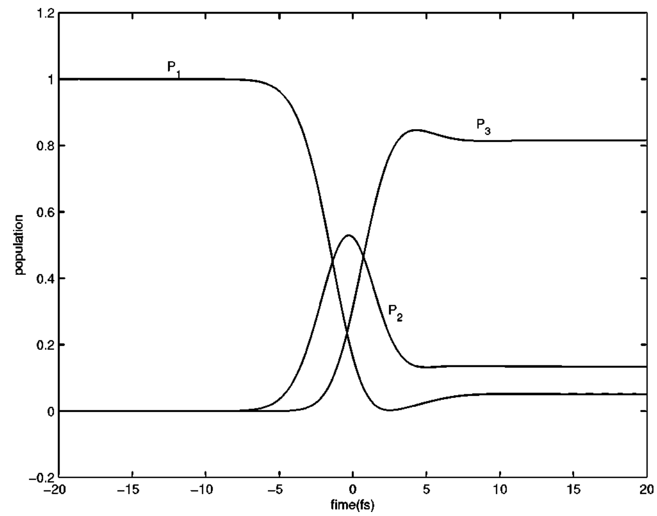

FIG. 3. Population evolution $P_n(t)$ for $n=1,2,3$ with the same parameters as in Fig. 2, but the rotating wave approximation is used in the calculation.

Figure 2 shows an example of the three-levels population evolution for $\gamma=1$. It can be seen that when the Rabi frequency is $\Omega=0.56$ fs^{-1} and the chirp rate is $\chi=0.04$ fs^{-2}, almost complete population transfer (98.9%) can be produced. Unlike the STIRAP and other adiabatic techniques, in this example, the population in the intermediate level $|2\rangle$, can reach large values. However, if the rotating wave approximation is used in the calculation, as shown in Fig. 3, the curves are very smooth and the final population of $P_3(t)$ decreases to 81.5%. These two figures clearly show the inapplicability of RWA in the case of few-cycle pulses interacting with three-level systems.

In Fig. 4, the final population of level $|3\rangle$, $P_3(\infty)$ is presented as the function of the Rabi frequency Ω. The curve shows the population transfer is robust to small Rabi-frequency changes. The maximal population transfer can be 98.9% and more than 90% population transfer can be achieved for $0.48<\Omega<0.62$.

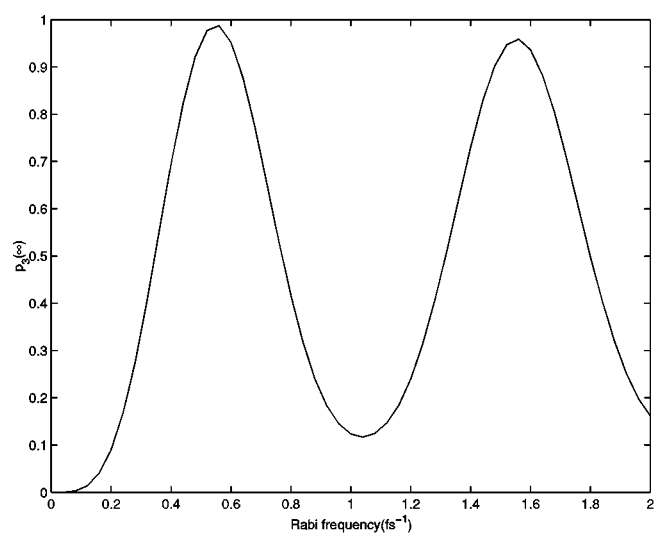

FIG. 4. Dependence of the final population transfer $P_3(\infty)$ with respect to Rabi frequency. $\chi=0.04$ fs^{-2} for $\tau=5$ fs and $\gamma=1$.

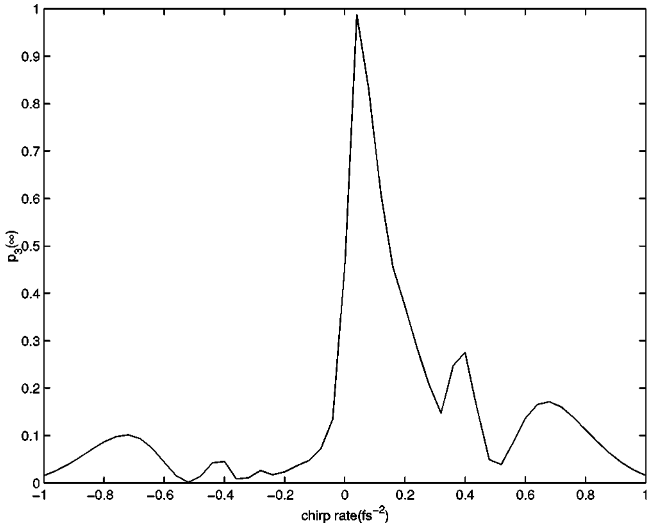

FIG. 5. Dependence of the final population transfer $P_3(\infty)$ with respect to chirp rate. $\Omega = 0.56\ \text{fs}^{-1}$, for $\tau = 5$ fs and $\gamma = 1$.

Also, we have plotted the dependence of $P_3(\infty)$ with respect to the chirp rate in Fig. 5. For opposite chirp rate, we see the population transfer is relatively small. Although, for positive chirp rate large population transfer can be obtained, the extent of the robustness is small.

To get the global behavior of population transfer, the contour map of $P_3(\infty)$ as the function of both the Rabi frequency and chirp rate is shown in Fig. 6. We find there are a certain region of Ω and χ in which large population transfer can be obtained. As these numerical results show, nearly complete population transfer is possible in the interaction of few-cycle pulse with three-level Λ atoms even for the pulse width as short as 5.9 fs under the condition $\gamma = 1$. But the population transfer is not very robust.

However, if the ratio of the dipole moments $\gamma \neq 1$, the maximum population transfer obtained will decrease.

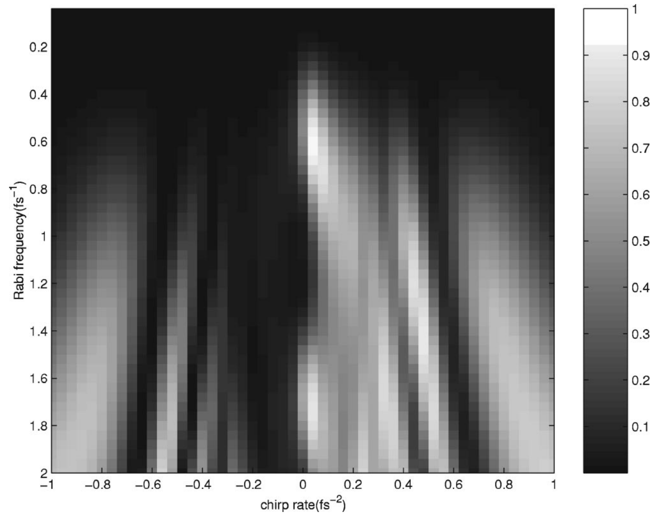

FIG. 7. Contour map of the final population transfer $P_3(\infty)$ for varying the Rabi frequency and varying chirp rate. $\tau = 5$ fs and $\gamma = 0.8$.

For $\gamma = 0.8$ and $\tau = 5$ fs, Fig. 7 gives the contour map of population $P_3(\infty)$. Compared with Fig. 6, the structure is qualitatively very similar, but the maximal population transfer is 96.2% when $\Omega = 0.6\ \text{fs}^{-1}$ and $\chi = 0.04\ \text{fs}^{-2}$, a little smaller than the maximum value for $\gamma = 1$.

If the ratio of the dipole moments decreases to $\gamma = 0.5$, as shown in Fig. 8, the maximal population transfer becomes much smaller. In this case, the value is 69.3%, when $\Omega = 0.72\ \text{fs}^{-1}$ and $\chi = 0.04\ \text{fs}^{-2}$.

The increase of the ratio of the dipole moments also decreases the achievable efficiency of population transfer. For $\gamma = 2.0$, the contour map of $P_3(\infty)$ is shown in Fig. 9. The maximal population transfer is 76.3%.

We have performed other simulations with different pulse width. For example, Fig. 10 gives the simulation results with

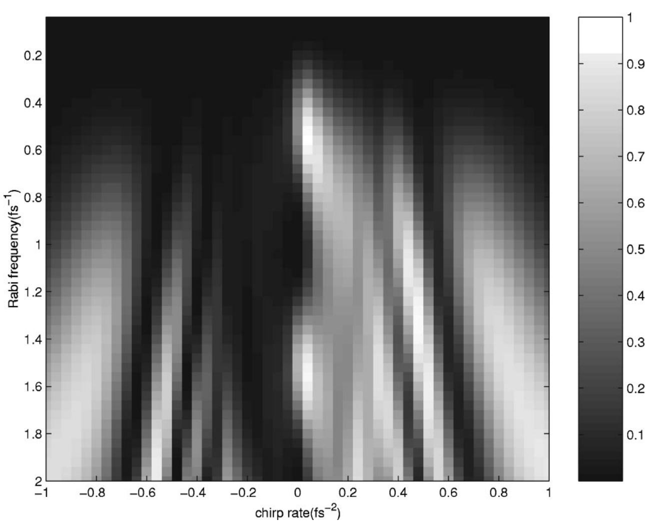

FIG. 6. Contour map of the final population transfer $P_3(\infty)$ for varying the Rabi frequency and varying chirp rate. $\tau = 5$ fs and $\gamma = 1$.

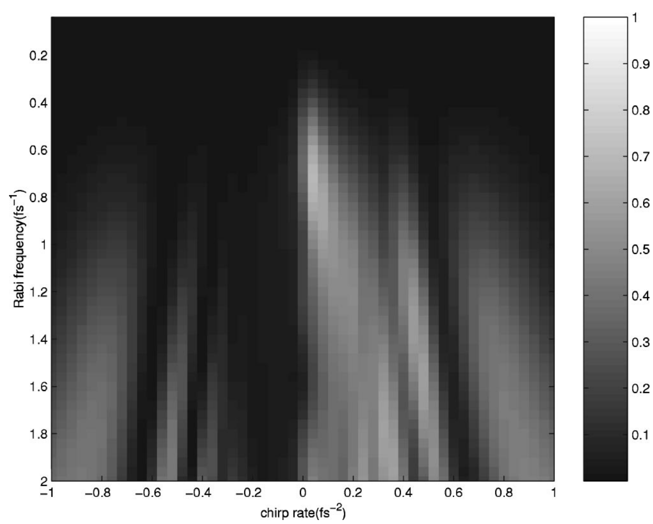

FIG. 8. Contour map of the final population transfer $P_3(\infty)$ for varying the Rabi frequency and varying chirp rate. $\tau = 5$ fs and $\gamma = 0.5$.

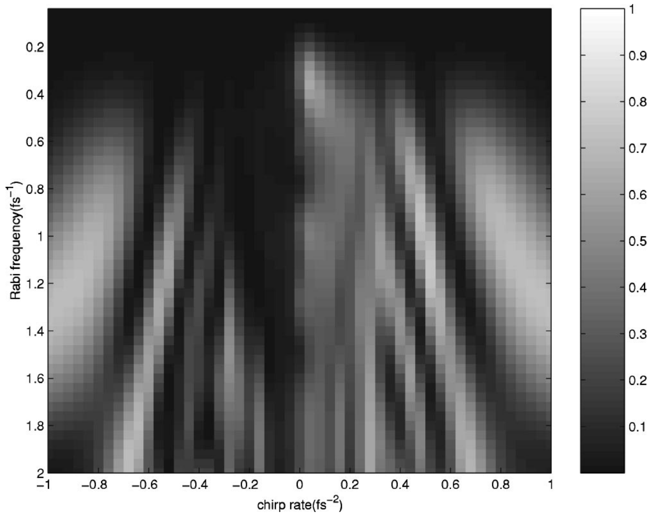

FIG. 9. Contour map of the final population transfer $P_3(\infty)$ for varying the Rabi frequency and varying chirp rate. $\tau=5$ fs and $\gamma=2$.

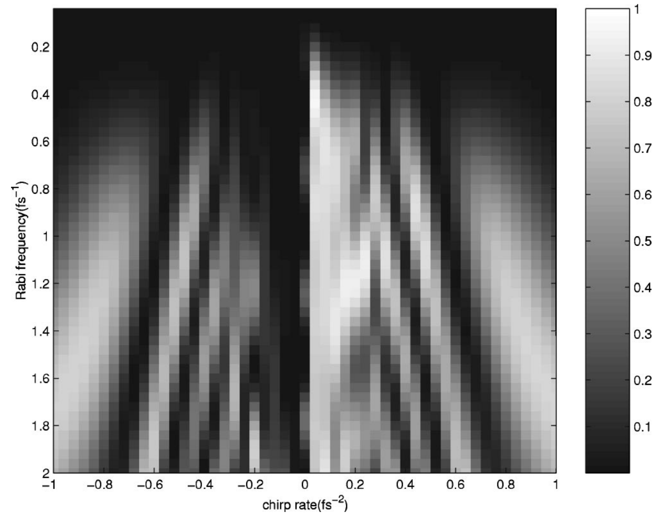

FIG. 10. Contour map of the final population transfer $P_3(\infty)$ for varying the Rabi frequency and varying chirp rate. $\tau=10$ fs and $\gamma=1$.

$\tau=10$ fs and $\gamma=1$. In this case, the maximal population transfer is 95.5%, when $\Omega=0.44$ fs^{-1} and $\chi=0.04$ fs^{-2}, smaller than the corresponding value with $\tau=5$ fs in Fig. 6.

From all these simulations we can get the following results:

(1) The interaction of few-cycle laser pulses with three-level atoms is much more complicated compared with multicycle pulses. The RWA cannot be used in this case and the different transitions can be stimulated simultaneously.

(2) The population transfer in the three-level atoms in a single few-cycle pulses laser is strongly dependent on the ratio of the dipole moments. If the ratio is near unity, almost complete population transfer can be obtained under appropriate values of the chirp rate and intensity. If the ratio deviates from unity largely, the maximal population transfer decreases seriously.

(3) The maximal population transfer has robustness in a small region of intensity and chirp rate.

In conclusion, the population transfer in the three-level atoms in a single few-cycle pulses laser is investigated in this paper with numerical simulation. The RWA is not used in the simulation. Almost complete population transfer can be achieved if the different transition dipole moments are almost the same and appropriate values of the intensity and chirp rate are selected.

This work was supported by the National Key Basic Research Special Foundation under Grant No. G1999075200.

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