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Population transfer by femtosecond laser pulses in a ladder-type atomic system*

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The population transfer in a ladder-type atomic system driven by linearly polarized sech-shape femtosecond laser pulses is investigated by numerically solving Schrödinger equation without including the rotating wave approximation (RWA). It is shown that population transfer is mainly determined by the Rabi frequency (strength) of the driving laser field and the chirp rate, and that the ratio of the dipole moments and the pulse width also have a prominent effect on the population transfer. By choosing appropriate values of the above parameters, complete population transfer can be realized.

Keywords: population transfer, femtosecond laser pulses, ladder-type system
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1. Introduction

There has been a growing interest in target population transfer[1,2] over the past decade, because of its very wide applications, such as, to chemical-reaction dynamics,[3] laser-induced cooling,[4] atomic optics,[5] the preparation of entanglement[6] and quantum computation.[7] There are many methods to achieve a population transfer in atomic and molecular systems. Stimulated Raman adiabatic passage (STIRAP) has emerged as a very efficient method to provide an almost complete population transfer,[8−10] but the requirement for large pulse areas seems to advise one against the use of short pulse lasers. Recent advances in ultrafast laser technology have made it possible to generate few-cycle and high intense pulses. Study of interaction between ultrashort laser pulse and atomic or molecular system has attracted much attention.[11−16] The use of the ultrashort pulse laser to obtain a population transfer may have some special benefits, such as, it is easy to realize the transition in the UV or VUV energy range, or control the level population distribution in the ultrafast process.[17] For pulses in the picosecond or sub-picosecond regime, adiabatic condition can be satisfied by chirping the frequency of the laser pulses, and nearly complete population transfer in atomic and molecular systems can be obtained. For example, Band and Magnes[18] studied the dynamics of chirped adiabatic passage by using temporally delayed pulses to selectively and fully transfer in a lambda-type three-level atomic system; Malinovsky and Krause[19] investigated the efficiency and the robustness of population transfer induced by intense, chirped laser pulses in diatomic molecules. Malinovsky and Krause[20] presented a general theory of adiabatic rapid passage caused by intense, linearly chirped laser pulses with a Gaussian profile and a fixed bandwidth and discussed the population transfer for two- and four-level systems. Most of these methods are based on the rotating wave approximation (RWA) and the adiabatic condition is required to be satisfied. However, the pulses in the picosecond or sub-picosecond regime contain only a few optical cycles of the carrier frequency and their spectrum width can reach several hundred nanometres. The RWA is not valid for the interaction of few-cycle pulses with atoms.[21,22] Cheng and Zhou[23] have studied the population transfer in a lambda-type three-level system driven by Gauss-shape femtosecond laser pulses in terms of the numerical calculation without including

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the RWA, and the maximal population transfer can reach 98.9%. Now we investigate an ultrafast population transfer in a ladder-type three-level atomic system driven by linearly polarized sech-shape femtosecond pulses by solving numerically the Schrödinger equation without including the RWA of the system, and the maximal population transfer can be 100%. In addition, we discuss the effects of Rabi frequency (strength) of the driving laser field, chirp rate, the ratio of the dipole moments and the pulse width on the population transfer, respectively.

2. Theoretical model

The three-level ladder-type atomic system considered here is shown in Fig.1. The transition frequency between the state $|2\rangle$ ($|3\rangle$) and state $|1\rangle$ ($|2\rangle$) is $\omega_1(\omega_2)$. The transition between levels 1 and 3 is electric dipole forbidden. Only one laser field interacts with the system. The free Hamiltonian is

$$H_0 = \mathrm{Diag}(0, \hbar\omega_1, \hbar\omega_1 + \hbar\omega_2). \tag{1}$$

The full Hamiltonian is

$$H(t) = H_0 - \boldsymbol{d} \cdot \boldsymbol{E}(t), \tag{2}$$

where $\boldsymbol{d}$ is the dipole moment operator, $\boldsymbol{E}(t)$ is the electric field of the laser and $\boldsymbol{E}(t) = \boldsymbol{e}E_0Q(t)\cos(\omega t - \chi t^2)$. Here $\boldsymbol{e}$ is the unit polarization vector, ω is the carrier frequency, χ is the chirp rate, E_0 is the maximal electric field amplitude, $Q(t)$ is the shape function and has the following form:[24]

$$Q(t) = \mathrm{sech}(t/\tau), \tag{3}$$

where τ is the parameter of the pulse width.

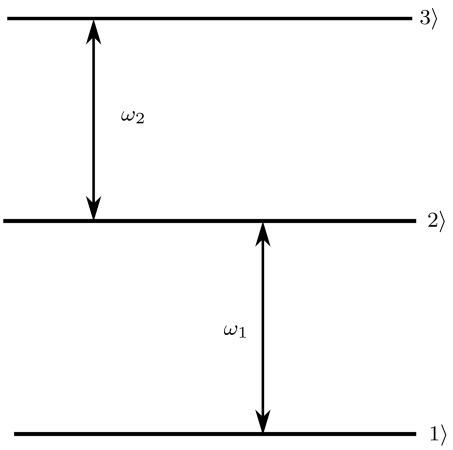


Fig.1. Tree-level ladder system.

We introduce the Rabi frequency $\Omega = \langle 1|\, \boldsymbol{d} \cdot \boldsymbol{e}\, |2\rangle\, E_0/\hbar$, and define the ratio of the transition dipole moments $\gamma = \langle 2|\, \boldsymbol{d}\cdot\boldsymbol{e}\, |3\rangle \,/\, \langle 1|\, \boldsymbol{d}\cdot\boldsymbol{e}\, |2\rangle$. The Hamiltonian now has the following matrix form:

$$H(t) = \begin{bmatrix} 0 & \hbar\Omega Q(t)\cos(\omega t - \chi t^2) & 0 \\ \hbar\Omega Q(t)\cos(\omega t - \chi t^2) & \hbar\omega_1 & \gamma\hbar\Omega Q(t)\cos(\omega t - \chi t^2) \\ 0 & \gamma\hbar\Omega Q(t)\cos(\omega t - \chi t^2) & \hbar(\omega_1 + \omega_2) \end{bmatrix}. \tag{4}$$

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

$$\mathrm{i}\hbar\frac{\mathrm{d}}{\mathrm{d}t}\begin{pmatrix} c_1 \\ c_2 \\ c_3 \end{pmatrix} = H(t)\begin{pmatrix} c_1 \\ c_2 \\ c_3 \end{pmatrix}. \tag{5}$$

In Eqs.(4) and (5), we have neglected the spontaneous emission from levels $|3\rangle$ and $|2\rangle$ since the interaction time is only a few cycles of the carrier frequency. Equations (4) and (5) are exactly quantum and the RWA is not used. Assume that at the initial time the system is in its ground level $|1\rangle$, i.e.

$$\rho_{11}(-\infty) = |c_1(-\infty)|^2 = 1,$$

$$\rho_{22}(-\infty) = |c_2(t)|^2 = 0,$$

and

$$\rho_{33}(-\infty) = |c_3(-\infty)|^2 = 0.$$

We will solve numerically the Schrödinger equation by using the eighth order Runge–Kutta method to investigate the population transfer.

3. Numerical result and discussion

In our simulation, the carrier frequency of the laser pulse is $\omega = 0.771\pi$ radian/fs corresponding to 778 nm central wavelength and $\tau = 5\,\mathrm{fs}$. The frequency detunings are $\omega_1 - \omega = -0.002\pi$ radian/fs and $\omega_2 - \omega = 0.002\pi$ radian/fs. Both transition frequencies are contained in the bandwidth of the pulse.

Figure 2 illustrates the populations as a function of the time for $\gamma = 1$. The parameters values are Rabi frequency $\Omega = 0.8\,\mathrm{fs}^{-1}$ and the chirp rate $\chi = 0.06\,\mathrm{fs}^{-2}$. From Fig.2 we can see that almost complete population transfer (99.7%) can be produced and the population in the immediate level $|2\rangle$ can reach a largest value when $t = 0$.

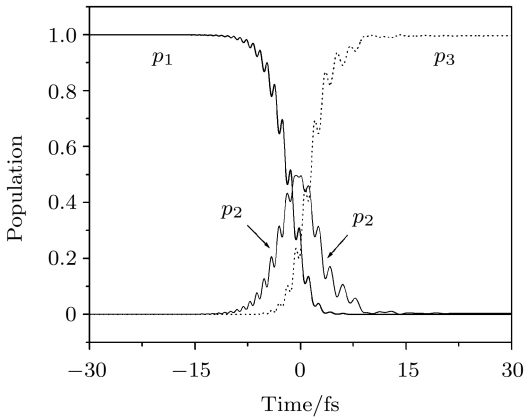


Fig.2. Population evolution $\rho_{ii}(t)$ for $i = 1, 2, 3$, with $\Omega = 0.8\,\mathrm{fs}^{-1}$, $\chi = 0.06\,\mathrm{fs}^{-2}$ for $\tau = 5\,\mathrm{fs}$ and $\gamma = 1$.

In Fig.3 the final population of level $|3\rangle$, $\rho_{33}(\infty)$, is presented as a function of Rabi frequency Ω. The other parameter values are the same as those in Fig.2. It is clear that the value of $\rho_{33}(\infty)$ equals the population transfer from the lowest level $|1\rangle$ to the highest level $|3\rangle$. The curve shows that when Rabi frequency is smaller, the population transfer increases rapidly with Rabi frequency increasing; when Rabi frequency is larger, the value of $\rho_{33}(\infty)$ nearly does not vary with Rabi frequency increasing and an almost complete population transfer (99.7%) can be produced.

Also, we plot the dependence of $\rho_{33}(\infty)$ on chirp rate χ in Fig.4, with the other parameter values being the same as those in Fig.2. From Fig.4 we can see that the population transfer is robust to small chirp rates and is symmetric about $\chi = 0$, and at $\chi = 0$, the population $\rho_{33}(\infty)$ takes a peak value, 0.8. With the value of $|\chi|$ increasing, $\rho_{33}(\infty)$ exhibits a non-periodic fluctuation. When $\chi = \pm 0.06$, the maximal population transfer can reach 99.7%. When $\chi = \pm 0.65$, then $\rho_{33}(\infty) = 0$, i.e. the population transfer does not occur.

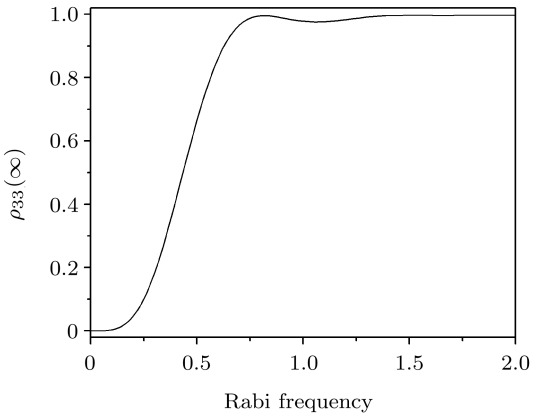


Fig.3. Dependence of the final population transfer $\rho_{33}(\infty)$ on Rabi frequency, with the other parameter values being the same as those in Fig.2.

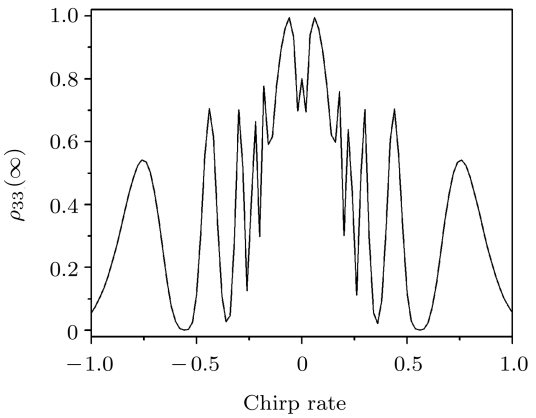


Fig.4. Dependence of the final population transfer $\rho_{33}(\infty)$ on chirp rate χ, with the other parameter values being the same as those in Fig.2.

In order to gain a deeper insight into the dependence of population transfer on Rabi frequency, chirp rate and ratio of the dipole moments γ, we give the contour maps of $\rho_{33}(\infty)$ as a function of both Rabi frequency and chirp rate for different values of γ in Figs.5, 6, 7 and 8, with the other parameter values being the same as those in Fig.2. The contour map of $\rho_{33}(\infty)$ as a function of both the Rabi frequency and chirp rate for $\gamma = 1$ is shown in Fig.5. From Fig.5 we can see that the distribution of population $\rho_{33}(\infty)$ is symmetric about $\chi = 0$; in the region of near $\chi = \pm 0.06$, the system keeps a large population transfer for any large value of Rabi frequency; when the value of $|\chi|$ is large, $\rho_{33}(\infty)$ first increases and

then decreases with Rabi frequency increasing; in addition, for some value of χ, $\rho_{33}(\infty)$ is always very small ($0 \le \rho_{33}(\infty) \le 0.1$) for any Ω; there are some regions of values of Ω and χ where a large population transfer ($0.9 \le \rho_{33}(\infty) \le 1$) can be obtained and the maximal population transfer can reach 100% (for example, when $\Omega = 1.665\,\mathrm{fs}^{-1}$ and $\chi = -0.1\,\mathrm{fs}^{-2}$).

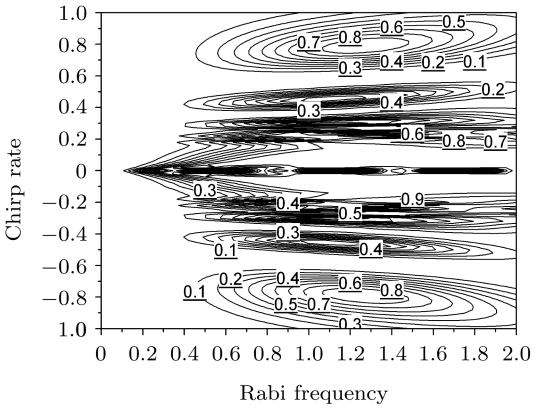


Fig.5. Contour map of $\rho_{33}(\infty)$, for Rabi frequency and chirp rate with $\tau = 5\,\mathrm{fs}$, $\gamma = 1$, and the other parameter values being the same as those in Fig.2.

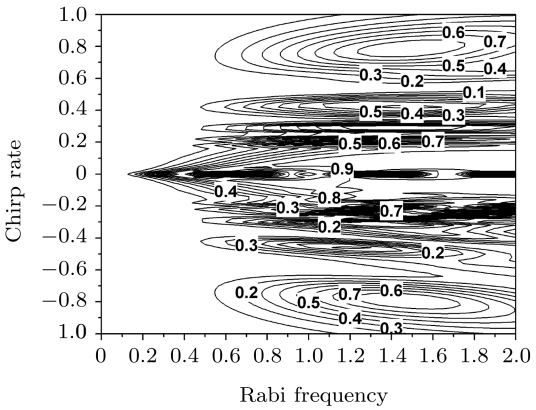


Fig.6. Contour map of $\rho_{33}(\infty)$ for Rabi frequency and chirp rate with $\tau = 5\,\mathrm{fs}$, $\gamma = 0.7$, and the other parameter values being the same as those in Fig.2.

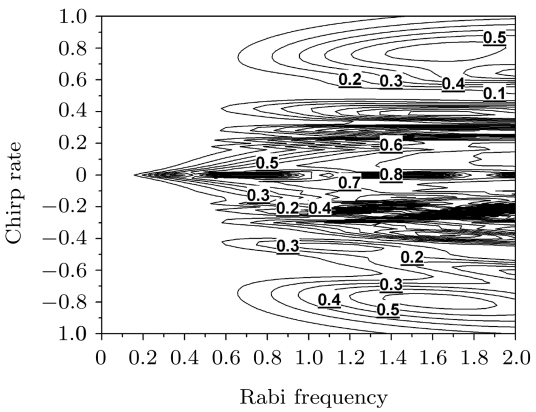


Fig.7. Contour map of $\rho_{33}(\infty)$ for Rabi frequency and chirp rate with $\tau = 5\,\mathrm{fs}$, $\gamma = 0.5$, and the other parameter values being the same as those in Fig.2.

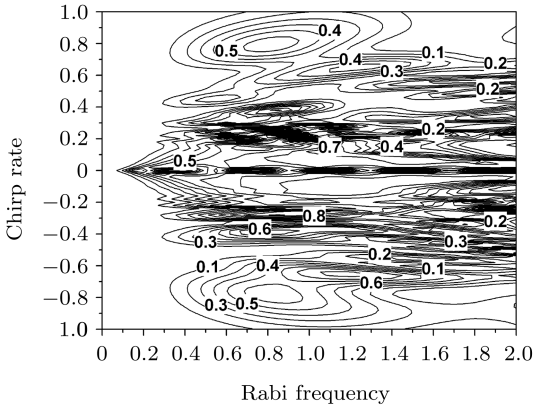


Fig.8. Contour map of $\rho_{33}(\infty)$ for Rabi frequency (τ) and chirp rate with $\tau = 5\,\mathrm{fs}$, $\gamma = 2$, and the other parameter values being the same as those in Fig.2.

If the ratio of the dipole moments $\gamma \neq 1$, the maximal population transfer and the region of large population transfer appearing on the $\chi - \Omega$ plane will decrease.

Figure 6 gives the contour map of $\rho_{33}(\infty)$ for $\gamma = 0.7$. Comparing Fig.6 with Fig.5, we find that Fig.6 is qualitatively similar to Fig.5, but the regions of larger population transfer decrease obviously; the population transfer is not symmetrical about $\chi = 0$ and the maximal population transfer for $\gamma = 0.7$ is 98.9%.

If we reduce the ratio of the dipole moments, γ, to 0.5 as shown in Fig.7, the region of large population transfer and the population transfer decrease again. And the maximal population transfer decreases to 88.9%.

The increasing of the ratio of the dipole moments also decreases the population transfer. We plot the contour map of $\rho_{33}(\infty)$ for $\gamma = 2$ in Fig.8. The maximal population transfer is only 83%.

We have performed other numerical calculations with different pulse widths. Figure 9 illustrates the numerical result, with $\tau = 10\,\mathrm{fs}$, $\gamma = 1$, and the other parameter values being the same as those in Fig.5. In this case, the maximal population transfer is 99.88%; however, comparing with Fig.5, the regions of $\rho_{33}(\infty)$ with larger values obviously reduce, and the distribution symmetry of the population $\rho_{33}(\infty)$ about $\chi = 0$ disappears.

Finally, it should be pointed out that our theoretical scheme can directly be applied to a real atomic system, i.e. rubidium atom, because the atomic level data used in the preceding discussion is taken from ^{87}Rb atom, and Fig.1 corresponds to ^{87}Rb atom level structure shown in Fig.10.

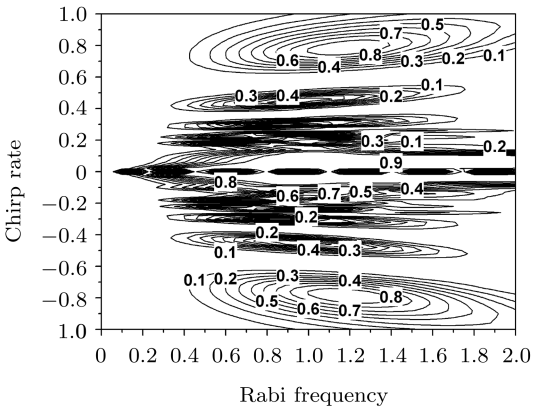


Fig.9. Contour map of $\rho_{33}(\infty)$ for Rabi frequency and chirp rate with $\tau = 10\,\text{fs}$, $\gamma = 1$, and the other parameter values being the same as those in Fig.2.

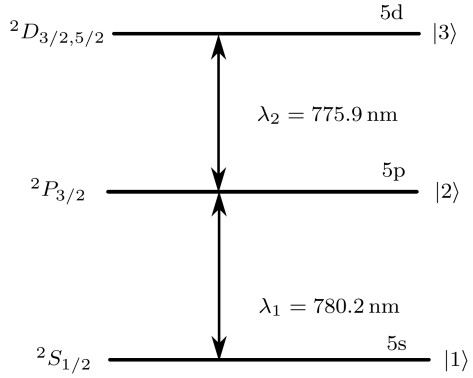


Fig.10. ^{87}Rb atom level structure.

4. Conclusions

We have investigated population transfer in a ladder-type three-level atomic system driven by linearly polarized sech-shape femtosecond pulses by solving numerically the Schrödinger equation without including the RWA. Our study shows that the population transfer from the lowest level to the highest level is mainly determined by the Rabi frequency of the driving laser field Ω and chirp rate χ, and that the ratio of the dipole moments γ and the pulse width τ also have a great influence on the population transfer. The maximal population transfer and the widest region, on the $\Omega - \chi$ plane, for a large population transfer appear when $\gamma = 1$. The region of large population transfer is wider when $\tau = 5\,\text{fs}$ than when $\tau = 10\,\text{fs}$. By choosing appropriate values of Ω, χ, γ and τ, a complete population transfer (100%) can be realized.

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