Coherent population transfer with chirped few-cycle laser pulses in an excited-doublet four-level system

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The behavior of population transfer in an excited-doublet four-level system driven by linear polarized few-cycle ultrashort laser pulses is investigated numerically. It is shown that almost complete population transfer can be achieved even when the adiabatic criterion is not fulfilled. Moreover, the robustness of this scheme in terms of the Rabi frequencies and chirp rates of the pulses is explored.

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The efficient excitation to particular states has attracted much attention in both physics and chemistry[1−10]. The stimulated Raman adiabatic passage technique for population transfer has been implemented with continuous or narrow band laser pulse (nanosecond time scale)[1], but the requirement for large pulse areas seems to advise against the use of short pulse lasers. However, the use of ultrashort laser pulse may have some distinct advantages, such as producing high energy frequencies, controlling level population within the time between collisions, and so on[2]. Recent studies have shown that short pulses can be used in coherent population transfer[3−5]. When few-cycle pulses are considered, a number of theoretical works have demonstrated the limitations of the rotating wave approximation (RWA) which is usually employed in the adiabatic passage[6−9].

Here, we investigate the coherent population transfer in an excited-doublet four-level system driven by two linear polarized chirped few-cycle laser pulses. By solving a time-dependent Schrödinger equation without the RWA, we demonstrate that almost complete population transfer in ultrashort scheme can be achieved even when adiabatic condition is not fulfilled. Moreover, we investigate the relationship among the maximal transfer efficiency, chirp rates, and the intensities of the pulses. It is found that in the ultrafast regime and beyond the adiabatic criteria, the scheme is very robust in terms of the variation of the Rabi frequencies and chirp rates when the pump and Stokes chirp rates have the opposite signs.

The excited-doublet four-level system is given in Fig. 1. It is a partial energy-level diagram for lithium atom, and we label the $2s$, $3s$, $5p$, and, $6p$ states $|1\rangle$, $|2\rangle$, $|3\rangle$, and $|4\rangle$, respectively. The 1—2 transition frequency is ω_{12}, the 4—2 transition frequency is ω_{42}, and the 3—2 transition frequency is ω_{23}. The transition between from level 1 to 4 and level 2 to 3 are electric dipole forbidden. The system parameters are deduced from the NIST tables: the transition frequency $\omega_{12} = 7.354$ fs^{-1}, $\omega_{42} = 2.226$ fs^{-1}, and $\omega_{23} = 0.259$ fs^{-1}[11]. We assume $\Omega_{p1}(t)$ and $\Omega_{p2}(t)$ are Rabi frequencies of the pump field coupling level 2 to 1 and level 3 to 1, respectively. $\Omega_{s1}(t)$ and $\Omega_{s2}(t)$ are Rabi frequencies of the Stokes field driving the transitions from level 2 to 4 and level 3 to 4. For simplicity, we choose $\Omega_p(t) = \Omega_{p1}(t) = \Omega_{p2}(t)$, and $\Omega_s(t) = \Omega_{s1}(t) = \Omega_{s2}(t)$, which can be written as

$$\Omega_{p,s}(t) = A_{p,s}(t)\cos(\omega_{p,s}t + 1/2\chi_{p,s}t^2), \quad (1)$$

where $\omega_{p,s}$ is the carrier frequency of pump and Stokes fields, $\chi_{p,s}$ is the linear chirp rate. $A_{p,s}(t)$ are the envelope of the pulse. Gaussian function is used as

$$A_{p,s}(t) = \Omega_{p0,s0}\exp(-t^2/\tau_{p,s}^2), \quad (2)$$

where $\Omega_{p0,s0}$ is the maximal electric field Rabi frequency and $\tau_{p,s}$ the parameter of the pulse width. Assuming that the pump and Stokes pulses are sufficiently smooth and have the same peak Rabi frequency $\Omega_{p0} = \Omega_{s0} = \Omega_0$ and the same pulse width $\tau_p = \tau_s = \tau$. For general stimulated Raman adiabatic passage technique, the adiabatic condition can be written as $\Omega_0\tau \gg 1$. In practical applications the pulse area should exceed 10 to provide efficient population transfer, $\Omega_0\tau > 10$[12]. In our results, the pulse areas are so small that the adiabatic condition need not be fulfilled.

Hamiltonian can be written as[13]

$$H(t) = \hbar\begin{pmatrix} -\omega_{12} & \Omega_p(t) & \Omega_p(t) & 0 \\ \Omega_p(t) & 0 & 0 & \Omega_s(t) \\ \Omega_p(t) & 0 & \omega_{23} & \Omega_s(t) \\ 0 & \Omega_s(t) & \Omega_s(t) & -\omega_{42} \end{pmatrix}. \quad (3)$$

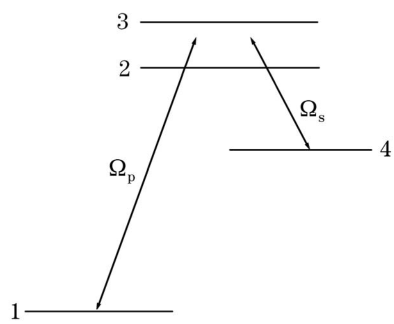

Fig. 1. The excited-doublet four-level system with two lower states $|1\rangle$ and $|4\rangle$ and excited-doublet states $|2\rangle$ and $|3\rangle$. The transition from the upper levels $|2\rangle$ and $|3\rangle$ to lower state $|1\rangle$ is driven by the pump pulse $\Omega_p(t)$; while the transition from the upper levels $|2\rangle$ and $|3\rangle$ to lower state $|4\rangle$ is driven by the Stokes pulse $\Omega_s(t)$.

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

$$i\hbar\frac{\mathrm{d}}{\mathrm{d}t}\begin{pmatrix} c_1 \\ c_2 \\ c_3 \\ c_4 \end{pmatrix} = H(t)\begin{pmatrix} c_1 \\ c_2 \\ c_3 \\ c_4 \end{pmatrix}. \quad (4)$$

We suppose the excited-doublet four-level system is in its ground level $|1\rangle$ at the initial time,

$$c_1(-\infty) = 1,\ c_2(-\infty) = 0,\ c_3(-\infty) = 0,\ c_4(-\infty) = 0.$$

By solving Eq. (4) without the RWA, we get the population distribution at $t \to +\infty$, $P_n = |c_n(+\infty)|^2$ $(n = 1, 2, 3, 4)$.

The laser parameters are the pulses width $\tau = 5$ fs (the full-width at half-maximum pulse width is 5.9 fs). We consider the case in which two-photo resonance is maintained: the pump pulse frequency $\omega_\mathrm{p} = 7.483$ fs^{-1}, and the Stoke pulse frequency $\omega_\mathrm{s} = 2.355$ fs^{-1}. The results to follow can, of course, be scaled to various laser and material parameters.

Figure 2 shows the computed population for states $|1\rangle$, $|2\rangle$, $|3\rangle$, and $|4\rangle$ as a function of time for $\Omega_0\tau = 4.0$ and chirp rates $\chi_\mathrm{p} = 0.20$ fs^{-2} and $\chi_\mathrm{s}\tau^2 = $ -0.22 fs^{-2}, respectively. It can be seen that nearly complete population transfer (99.7%) can be produced. Unlike the adiabatic passage techniques, populations of P_2 and P_3 during intermediate time can reach large values ($P_2 = 40.8\%$, $P_3 = 17.3\%$) because the adiabatic criteria is not fulfilled in this case. However, in spite of the intermediate process, level $|4\rangle$ receives the most population and the population in any other level never exceeds 0.3% at the final time of the pulses. When few-cycle laser pulses are considered, the individual carriers will have much profound effect, and time-derivative-driven nonlinearities will have a significant impact on the interaction of laser pulses with nonlinear materials which can lead to strong oscillation features during the evolutions of population[6]. These features are not present in RWA solutions.

Moreover, we investigate the relationship between the final population of level $|4\rangle$ and the chirp rates. In Fig. 3, the final population of level $|4\rangle$ ($P_4(\infty)$) is presented as the function of chirp rates χ_p and χ_s when the Rabi frequencies are relatively small ($\Omega_0 = 0.8$ fs^{-1}, i.e., $\Omega_0\tau$ = 4.0). In the contour map, it can be seen that the population transfer is robust to the magnitude of the chirp rates and almost all high transfer efficiencies (larger than 90%) can be obtained in the area where the pump and the Stokes chirp rates have the opposite signs. However, when the pump and Stokes chirp rates are the same signs, population transfer can hardly be achieved.

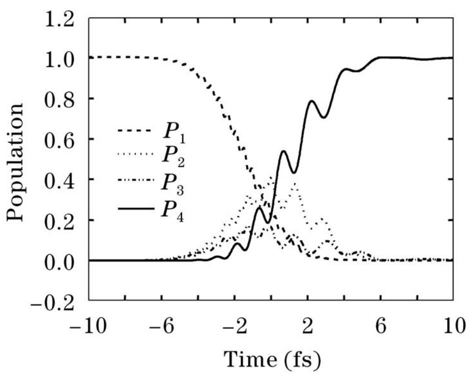

Fig. 2. Population evolution $P_n(t)$. $\Omega_0\tau = 4.0$, χ_p =0.2 fs^{-2} and $\chi_\mathrm{s} = -0.22$ fs^{-2} for $\tau = 5$ fs.

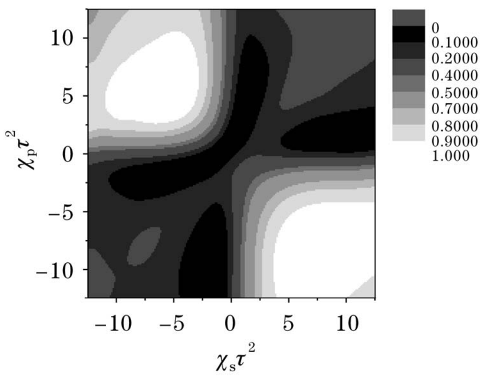

Fig. 3. Contour map of the final population transfer $P_4(\infty)$ for varying pump chirp rates and Stokes pulses. $\Omega_0\tau = 4.0$.

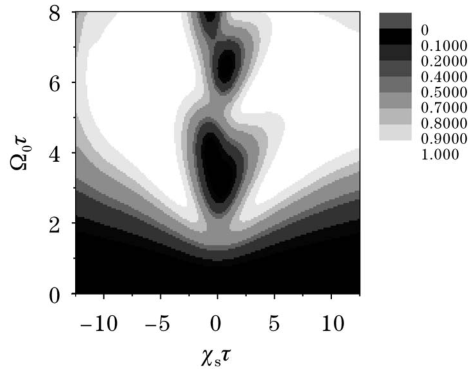

Fig. 4. Contour map of the final population transfer $P_4(\infty)$ for varying the chirp rates and the Rabi frequencies when the Stokes and pump chirp rates have the opposite sign. $\chi_\mathrm{p}\tau^2 = -\chi_\mathrm{s}\tau^2$.

Further consideration is given to the case in which the final population of level 4 ($P_4(\infty)$) is presented as the function of chirp rates and the Rabi frequencies when the pump and Stokes chirp rates have the opposite signs ($\chi_\mathrm{p}\tau^2 = -\chi_\mathrm{s}\tau^2$). As shown in Fig. 4, the pulse areas are so small that the adiabatic condition can not be fulfilled. However, the scheme is very robust to both the chirp rates and the Rabi frequencies. Almost complete population transfer can be achieved in this case. Hence, in the ultrashort regime, controlling chirping is a much more robust method for population transfer and is easy to be implemented in experiments.

The qualitative explanation for the mechanism of such scheme is similar to Refs. [12] and [14], where population transfer with chirped laser pulse in molecular system was explained. At the beginning of the evolution, the pump and Stokes pulses are on two-photon resonance with the desired transition frequency. For this scheme, the most important feature of modulated pulses is the opposite sign chirp rates, which describes the temporal variation of pulse frequencies. For properly high optical intensity, Rabi oscillation can cycle the population to target state. By the time the Rabi oscillation starts to cycle the population of target state back to the initial state, the pulses frequencies have become far away from resonant due to the opposite sign chirp rates. Hence, the population can be trapped on the target state in this case. Whereas, when the pump and Stokes chirp rates have the same signs, it can not become quickly off-resonant with the transition frequency and certain electronic population of target state can be cycled back to the ground state. Therefore, complete population transfer can be difficult to achieve when pump and Stokes chirp rates have the same signs. All these analyses are consistent with our numerical results (see Fig. 3).

In conclusion, we have investigated the coherent population transfer in an excited-doublet four-level system driven by two linear polarized chirped few-cycle laser pulses. By solving a time-dependent Schrödinger equation without the RWA, we have demonstrated that almost complete population transfer can be achieved when the Rabi frequencies are relative small and adiabatic condition can not be fulfilled. Moreover, we have investigated the relationship among the maximal transfer efficiency, chirp rates, and the intensities of the pulses. It was found that in the ultrashort regime and beyond the adiabatic condition, the scheme is very robust in terms of the variation of the Rabi frequencies and chirp rates when the pump and Stokes chirp rates have the opposite signs.

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