Ultrafast population transfer in a Λ-configuration level system driven by few-cycle laser pulses*

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The feasibility of population transfer from a populated level via an intermediate state to the target level driven by few-cycle pulses is theoretically discussed. The processes of on- or far-resonance stimulated Raman scattering with sequential or simultaneous ultrashort pulses are investigated respectively. We find that the ultrashort pulses with about two optical cycles can be used to realize the population operation. This suggests that the population transfer can be completed in the femtosecond time scale. At the same time, our simulation shows that the signal of the carrier-envelope-phase-dependent effect can be enlarged due to quantum interference in some conditions. Our theoretic study may promote the research on the coherent control via ultrashort pulses in the related fields.

Keywords: ultrafast processes, coherent control of atomic interactions with photons, quantum state engineering and measurements

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1. Introduction

The coherent population transfer manipulation of quantum systems by interaction with coherent radiation fields has become an important research topic in the field of quantum optics. The stimulated Raman transition is an extremely powerful tool for laser manipulation of atoms and molecules. The two delayed but partially overlapping laser pulses which drive a three-state Λ system can be used to transfer the population adiabatically from the initially populated state to the target state, which is called stimulated raman adiabatic passage (STIRAP) and was discovered and developed by Bergmann and co-workers,[1–3] which is known as a fast and efficient tool for population transfer. STIRAP can be used to create coherent superposition of states in quantum systems.[4–10] Meanwhile, STIRAP has found a huge number of applications in atomic and molecular physics, such as the laser-assisted isomerization process,[11] selective dissociative in molecular systems,[12,13] and Raman photoassociation for creating a coherent molecular Bose–Einstein condensate (BEC) from an atomic BEC.[14–16] In short, the related population manipulation has become a very popular technique for coherent population transfer in quantum systems.

Recently, much attention has again been paid to STIRAP, even though it has been studied for over twenty years. For example, many new types of pulses, such as time-separated chirped pulses and a train of counter intuitively ordered pump-Stokes pulse pairs,[17–21] were applied to transfer the population of atomic and molecular states. Reference [22] proposed an optimization of the fidelity of the STIRAP technique with optimum pulse shapes. References [23] and [24] detailed an alternative efficient method of population transfer where sequential pulses are used with a large one-photon detuning. Unlike the STIRAP, for which the dynamics adiabatically projects along a dark state, the intuitive process follows a bright state. It has been named bright STIRAP (b-STIRAP).[25,26] It should be emphasized that for all the above schemes population transfer needs two long pulses at least. Wherein the full-width at half-maximum (FWHM) of the pulses contains many cycles of laser oscillation. At the same time, femtosecond laser pulses have been proposed to realize the coherent population transfer. Different mechanisms to produce a superposition of atomic and molecular states by ultrashort laser pulses were proposed theoretically.[27–31] In addition, population transfer to the excited electronic state of Li_2 by transform limited delayed femtosecond pulses has been investigated in experiment.[32] With the progress in the generation of femtosecond laser pulses,[33,34] a new class of phase-dependent phenomena induced by the carrier-envelope phase (CEP) of a laser pulse consisting of a few cycles of electromagnetic field were explored.[33,35,36] For such cases, the ultrashort pulses can be so strong that the extra-unclear orbital electrons in atoms and molecules can be ionized. It should be noticed that the CEP is the absolute phase of the laser pulse and the single pulse interacting with matter can exhibit

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the phase dependence on many physical processes.[33] Up to now, the studies of few-level systems interacting with such ultrashort laser pulses have focused on manifestations of the CEP.[37–41] For example, a quantum beat signal[37] induced in a three-level system, four-level atomic interferometer,[40] and the population of quantum states of polar molecules.[41] As a consequence, the study on the phase dependent effects in optical physics has become a hot topic and further research will help us to deepen the understanding of coherent control. The application of ultrashort laser pulses on coherent population transfer has important practical significance. For example, all the processes can be completed within the femtosecond time scale, in which there is no need to regard all of the decay effect in the atomic and molecular systems. There are two difficulties in the population operation realized by ultrashort pulses: several excited states are coupled simultaneously due to the pulses with a broad spectrum; the fast-rotating term in the Hamiltonian observably affects the population transfer in the strong coupling regime. The question is that whether or not the ultrashort pulse can be applied in the population transfer. If the ultrashort pulses can work, it is natural to ask how short pulses and which quantum systems can be used.

In the present paper, we focus on the coherent population transfer with an ultrahigh speed based on the few-cycle pulses. We theoretically investigate the population dynamics in a three-level atom driven by few-cycle laser pulses and discuss the potential applications of ultrashort pulses in the related fields. This article is structured as follows. We first describe, in Section 2, the Raman transition in a three-level system driven by two few-cycle pulses and we show that this scheme can be used to effectively transfer the population to the target state. We then derive, in Section 3, expressions for the behavior of the quantum-mechanical amplitudes in the approximation. In Section 4, we show the conclusion.

2. Model and numerical simulation

The system under investigation is a three-level Λ-configuration quantum system which is driven by two ultrashort laser pulses with different polarizations as illustrated in Fig. 1(a). Here, we adopt a semiclassical model in which the quantum system interacts with a classical electric field. We label the states of the system by $|n\rangle$, with states $|1\rangle$ and $|3\rangle$ coupled by the pump field $E_{\rm p}$, and $|2\rangle$ and $|3\rangle$ coupled by the "Stokes" field $E_{\rm c}$, respectively. It should be pointed out that such a three-level quantum system interacting with ultrashort pulses is practical. The atomic and molecular systems usually are not suitable because the corresponding dipole momentums are too small and the intensities of ultrashort pulses are too strong, in which the ionization rate should be considered. Recently, many researchers reported that some artificial atomic systems, such as quantum dots[42] and quantum wells,[43] can be applied to investigate the dynamics in the strong coupling regime. One alternative experiment system is symmetric coupled double quantum wells consisting of two 5.09-nm-wide GaAs wells, separated by a 1.13-nm-wide pure AlAs central barrier, and sandwiched between $Al_{0.3}Ga_{0.7}As$ layers.[44] In such a system, there are only four bound states, as shown in Fig. 1(b). After an amount of numerical simulations, we find that the two excited states can be regarded as one level when $2\pi\hbar/\varepsilon \gg \tau$. Here, ε and τ are the energy gap of the two-excited states and the few-cycle pulse duration, respectively. In Appendix A, some numerical results are given to demonstrate this point. Another alternative system is a symmetric 8 nm–4 nm–8 nm $GaAs/Al_{0.33}Ga_{0.67}As$ coupled quantum well.[45] In fact, some three-level models have been used to describe the interaction in the femtosecond time scale.[32,46–51] In conclusion, a practical three-level model can be obtained to study a quantum system interacting with two ultrashort pulses with different polarization in some conditions.

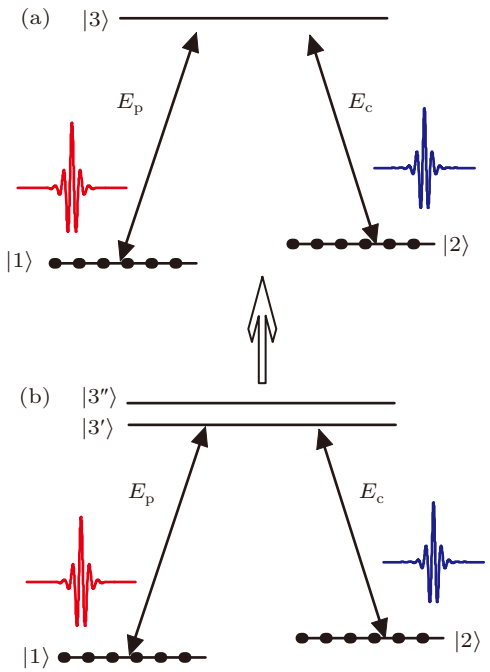


Fig. 1. (color online) Schematic diagram of the considered quantum systems. (a) Three-level Λ-type system; (b) four-level double-Λ-type system. The ground states $|1\rangle$ and $|2\rangle$ are coupled to the excited state(s) $|3\rangle$ ($|3'\rangle$ and $|3''\rangle$) by ultrashort laser pulses $E_{\rm p}$ and $E_{\rm c}$, respectively. If the energy gap ε of states $|3'\rangle$ and $|3''\rangle$ is small enough, such as $2\pi\hbar/\varepsilon \gg \tau$ (τ is the ultrashort pulse duration), the four-level system can be simplified as a three-level system, as shown in (a).

Taking the eigenfunctions $|1\rangle$, $|2\rangle$, $|3\rangle$ as a convenient basis set for the description of the population transfer process, the time-dependent Schrödinger equation of the corresponding probability amplitudes $\boldsymbol{C}_i$ for such a three-level system in the interaction picture can be archived as follows in matrix form:

$$\dot{\boldsymbol{C}}(t) = -\frac{\mathrm{i}}{\hbar}\boldsymbol{H}\boldsymbol{C}(t), \tag{1}$$

where $\boldsymbol{C} = (\boldsymbol{C}_1,\ \boldsymbol{C}_2,\ \boldsymbol{C}_3)^{\mathrm{T}}$ and the evolution matrix $\boldsymbol{H}$ has the form

$$\boldsymbol{H} = \begin{pmatrix} 0 & 0 & \mathrm{e}^{-\mathrm{i}\omega_{31}t}\Omega_{\mathrm{p}} \\ 0 & 0 & \mathrm{e}^{\mathrm{i}\omega_{21}t-\mathrm{i}\omega_{31}t}\Omega_{\mathrm{c}} \\ \mathrm{e}^{\mathrm{i}\omega_{31}t}\Omega_{\mathrm{p}} & \mathrm{e}^{\mathrm{i}\omega_{31}t-\mathrm{i}\omega_{21}t}\Omega_{\mathrm{c}} & 0 \end{pmatrix}.$$

Here, ω_{21} and ω_{31} are the transition frequencies of transition $|1\rangle \leftrightarrow |2\rangle$ and $|1\rangle \leftrightarrow |3\rangle$, respectively, $\Omega_{\mathrm{p,c}} = x_{31,32}E_{\mathrm{p,c}}(t)/\hbar$ is the Rabi frequency of the pump (Stokes) laser field, and $x_{31,32}$ is the dipole matrix moment $\langle 3|e\boldsymbol{x}|1,2\rangle$. In the above derivation, we have set $x_{31,32} = x_{13,23}$ because the dipole phases do not impact the dynamics of the system

$$E_{\mathrm{p}}(t) = E_{\mathrm{p}}^0 \exp\left[-4\ln 2\left(\frac{\omega_{\mathrm{p}}t}{2\pi N_{\mathrm{p}}}\right)^2\right]\cos(\omega_{\mathrm{p}}t + \phi_{\mathrm{p}})$$

and

$$E_{\mathrm{c}}(t) = E_{\mathrm{c}}^0 \exp\left\{-4\ln 2\left(\frac{\omega_{\mathrm{c}}(t-\Delta T)}{2\pi N_{\mathrm{c}}}\right)^2\right\}\cos[\omega_{\mathrm{c}}(t-\Delta T)+\phi_{\mathrm{c}}],$$

with the time delay ΔT are the Gaussian fields, $\omega_{\mathrm{p,c}}$ and $\phi_{\mathrm{p,c}}$ denote the carrier frequency and the CEPs, and $N_{\mathrm{p,c}}$ is the number of the cycles for the full width at half maximum (FWHM) of the amplitudes of pump (Stokes) laser fields. There is another important parameter $A_{\mathrm{p,s}} = \int_{-\infty}^{+\infty} E_{\mathrm{p,c}}^0 \exp[-4\ln 2(\omega_{\mathrm{p,c}}t'/2\pi N_{\mathrm{p,c}})^2]\,\mathrm{d}t'$, which means the area of the corresponding pulse envelope. If we assume that initially only one state was populated in a two-state system driven by a pulse, it is interesting that this transition probability vanishes when A is an even integer multiple of π. For an odd integer multiple of π, we get complete population inversion while the half-integer multiple of π gives an equal coherent superposition of the initial and the final states.

In general, the rotating wave approximation is invalid for the interaction between the few-level system and the ultrastrong (or ultrashort) pulses. Then, the convenient slowly varying Hamiltonian can not be yielded in our system. Although equation (1) is a set of linear first-order partial differential equations with time-varying coefficients, it is difficult to obtain an exact and analytical solution. The time evolution of the population can be obtained by means of a fourth-order Runge–Kutta method. In the following contents, we make numerical simulations to investigate the population dynamics in the three-level system driven by ultrashort pulses. During the process of numerical calculations, we use atomic units in which $e = \hbar = 1$.

We shall now begin to investigate the time evolution of the population on the three states coupled by an intuitive pair of few-cycle laser pulses under the initial conditions $\boldsymbol{C}_1(t_0) = 1$ and $\boldsymbol{C}_2(t_0) = \boldsymbol{C}_3(t_0) = 0$, which is shown in Fig. 2. Here, the analogical case of STIRAP is considered, such as the two-photon Raman resonant process. The few-cycle pulses that contain 2 cycle number in the FWHM. The few-cycle pulse's profile is shown in a read curve in the inset of Fig. 2(a). The CEPs of the pump and Stokes laser fields are fixed to be zero and the time delay is different. From these plots, we can find that the intuitive pair of few-cycle laser pulses with small time delay can transfer almost all the population from state $|1\rangle$ to $|2\rangle$ (see Fig. 2(b), the error rate is about 1%). The transfer efficiency can be enhanced by increasing the pulse duration. When the time delay is enlarged, the efficiency of population transfer will be worse (see Figs. 2(c) and 2(d), the error rate is about 6%). As we know, a level crossing process appears when the system is driven by an high enough intensity field, which will induce Landau–Zener transition.[52] The moment of Landau–Zener transition corresponds to the region where the population of state $|1\rangle$ falls fastest. This process occurs in most of our simulations. Due to this mechanism, the population of state $|1\rangle$ is transferred rapidly to state $|2\rangle$ via the excited state $|3\rangle$. In our case, the strength of the applied laser field is strong enough that a slight Landau–Zener transition occurs. The rapid decline (increase) regimes in the population evolution are the corresponding signals. Our results show that the ultrashort laser pulses with small time delay can be applied as coherent population transfer. In our numerical simulation, the ultrashort pulse consists of two optical cycles. Moreover, it should be mentioned that the value of pulse area which can transfer total population to the target state depends on the delay time (inspect Fig. 2) and the population transfer process is non-adiabatic.

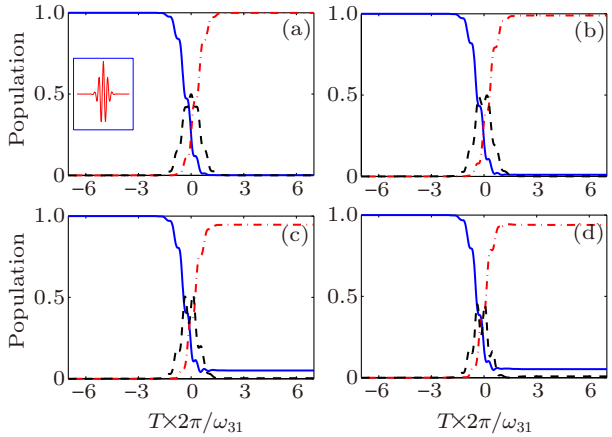


Fig. 2. (color online) Time evolution of the population on the three states coupled by an intuitive pair of few-cycle laser pulses with the same duration but different time delay. This process is similar to one of STIRAP. The solid, dash-dot, and dashed lines depict the population dynamics of states $|1\rangle$, $|2\rangle$, and $|3\rangle$, respectively. (a) $\Delta T = 0$ and $x_{31,32}E_{\mathrm{p,c}}^0/\hbar = 0.3321\omega_{31}$ corresponding to $\sqrt{2}\pi$; (b) $\Delta T = -0.2\pi/\omega_{31}$ and $x_{31,32}E_{\mathrm{p,c}}^0/\hbar = 0.3562\omega_{31}$ corresponding to $\sqrt{2.30}\pi$; (c) $\Delta T = -0.4\pi/\omega_{31}$ and $x_{31,32}E_{\mathrm{p,c}}^0/\hbar = 0.3713\omega_{31}$ corresponding to $\sqrt{2.50}\pi$; (d) $\Delta T = -0.6\pi/\omega_{31}$ and $x_{31,32}E_{\mathrm{p,c}}^0/\hbar = 0.3823\omega_{31}$ corresponding to $\sqrt{2.65}\pi$. Other parameters are: $\phi_{\mathrm{p}} = 0$, $\phi_{\mathrm{c}} = 0$, $\omega_{\mathrm{p}} = \omega_{\mathrm{c}} = \omega_{31} = \omega_{32}$ and $N_{\mathrm{p,c}} = 2$. The curve of a ultrashort pulse where the CEP and cycle number are equal to 0 and 2 is shown in the subplot of (a).

In following, we will check the CEP's impacts on the population dynamics in detail. For the few-cycle pulses, it

is the oscillating electric field itself rather than the envelope that drives the interaction. Figure 3 shows that the time evolution of the population driven by two simultaneous ultrashort pulses whose duration contains one oscillation cycle. Such a pulse is denoted as the curve (red solid line) in the inset of Fig. 3(a). As expected, the remarkable CEP-dependent phenomena occurs. In fact, we calculate the population dynamics of the three-level system driven by only one such ultrashort pulse. We find that the impact of CEPs on the final population is very weak, which is very different from the case in Fig. 3. In some sense, the signal of the CEP-dependent effect is enlarged owing to the quantum interference when the pulses are applied to the system simultaneously. With the increase of the pulse duration, the impact of CEPs will be suppressed. For comparison, figure 4 depicts the same case as Fig. 3 but for that the FWHM cycle number is equal to 2. Inspecting these two figures, we can find that the CEP effect is slight even for the pulse with two cycles in its FWHM. When the pulse duration increases further, the effect of CEP can be absolutely ignored and the rotating-wave approximation can be used in such an intensity regime. Here, we can see that the pulses with two optical cycles can be used to carry out our population operation.

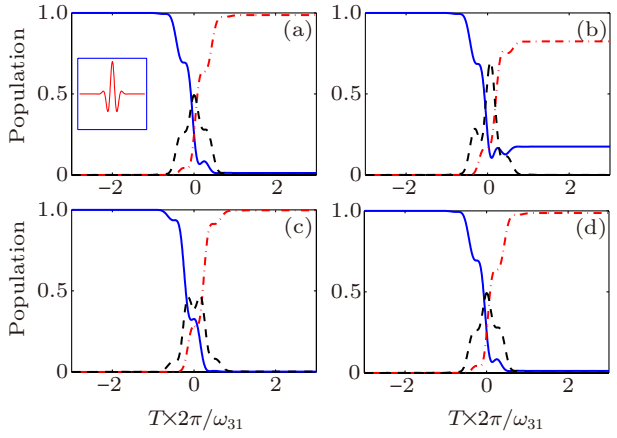


Fig. 3. (color online) Time evolution of the population on the three states coupled by two simultaneous few-cycle laser pulses with the same envelope but different CEPs. The solid line, dash-dot line, and dashed line in the above plots depict the population dynamics of states $|1\rangle$, $|2\rangle$, and $|3\rangle$, respectively. (a) $\phi_p = 0$ and $\phi_c = 0$; (b) $\phi_p = 0$ and $\phi_c = 0.5\pi$; (c) $\phi_p = 0.5\pi$ and $\phi_c = 0.5\pi$; (d) $\phi_p = 0$ and $\phi_c = \pi$. Other parameters: $\omega_p = \omega_c = \omega_{31} = \omega_{32}$, $\Delta T = 0$, $N_{p,c} = 1$, $x_{31,32}E^0_{p,c}/\hbar = 0.6643\omega_{31}$ corresponding to $\sqrt{2}\pi$. The curve of a ultrashort pulse where the CEP and cycle number are equal to 0 and 0.5 are shown in the inset of (a).

In addition, we should emphasize that the efficiency of the rotating-wave approximation relies not only on the pulse duration but also on the amplitude strength of the pulses. In order to demonstrate this point, the population dynamics with stronger pulses is shown in Fig. 5. Here, we set that the parameters are the same but the amplitudes of these two pulses are three times of the one in Fig. 4. From Fig. 5, we can observe the significant CEP-dependent phenomena again. At the same time, we find that the moment when the level crossing happened is different for the pulses owing to the different CEPs [see Figs. 3 and 5]. Due to this reason, the CEP effect can be observed easily, which is helpful for us to understand the strong nonlinear optical process. As a conclusion, the significant CEP-dependent phenomena depend on the strength of the amplitude and the slope of the electric field curve, which is yielded by the property of Landau–Zener tunneling.

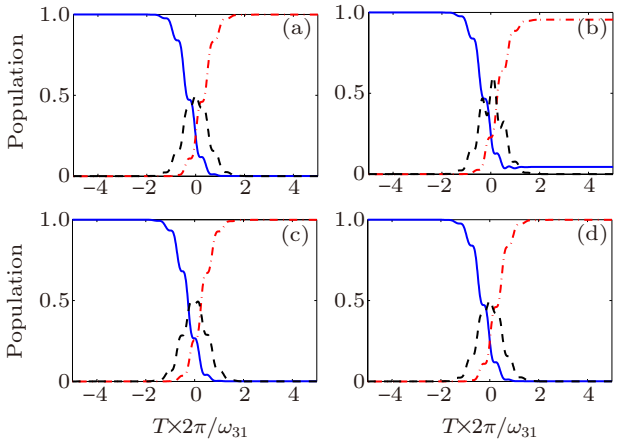


Fig. 4. (color online) Time evolution of the population on the three states coupled by two simultaneous few-cycle laser pulses with the same envelope but different CEPs. (a) $\phi_p = 0$ and $\phi_c = 0$; (b) $\phi_p = 0$ and $\phi_c = 0.5\pi$; (c) $\phi_p = 0.5\pi$ and $\phi_c = 0.5\pi$; (d) $\phi_p = 0$ and $\phi_c = \pi$. Other parameters: $\omega_p = \omega_c = \omega_{31} = \omega_{32}$, $\Delta T = 0$, $N_{p,c} = 2$ and $x_{31,32}E^0_{p,c}/\hbar = 0.3321\omega_{31}$ corresponding to $\sqrt{2}\pi$.

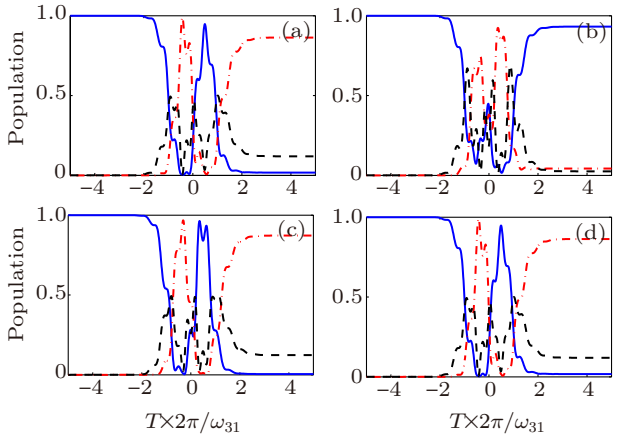


Fig. 5. (color online) Time evolution of the population on the three states coupled by two simultaneous few-cycle laser pulses with the same envelope but different CEPs. Other parameters: $\omega_p = \omega_c = \omega_{31} = \omega_{32}$, $\Delta T = 0$, $N_{p,c} = 2$, and $x_{31,32}E^0_{p,c}/\hbar = 0.9962\omega_{31}$ corresponding to $\sqrt{18}\pi$. Here, the intensity of the laser pulses is increased.

After analyzing an amount of numerical results about the impact of CEPs on the population dynamics, we obtain that ϕ_p and ϕ_c are two independent parameters. This point is different from the characterization of the regular interference cases, in which the observable process depends on the difference of the corresponding phases. In order to illustrate this, the impacts of the CEPs on the population dynamics of the target states $|2\rangle$

at different time moments are shown in Fig. 6. In that simulation, the pulse duration is the same as the one in Fig. 5. The study of such phenomena and the corresponding technology exploration have potential applications in the CEP measurement for the weak-field regime (without ionization process). In addition, it should be noticed that the area of the two pulses is equal to $\sqrt{2}\pi$ which can make the population completely transfer from state $|1\rangle$ to state $|2\rangle$. In order to maintain structural integrity, the reason for total population transfer via such a $\sqrt{2}\pi$ pulse will be discussed in Section 3. Until now, we have investigated the coherent population transfer of the system driven by two simultaneous ultrashort pulses in different conditions and there are significant CEP effects in some conditions.

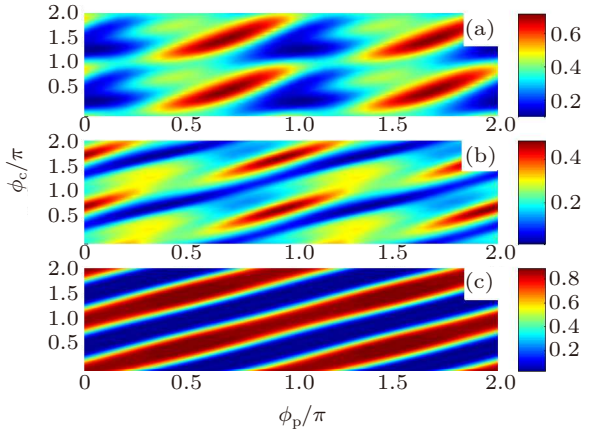


Fig. 6. (color online) Impact of CEPs on the population on the target states $|2\rangle$ at different time moments. The parameters are the same as these in Fig. 5 but the CEPs are varying. (a) $\Delta T = 0$; (b) $\Delta T = 1$; (c) $\Delta T = 4$.

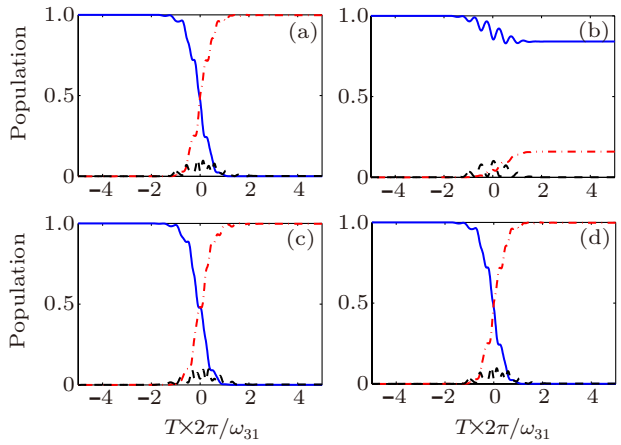


Fig. 7. (color online) Time evolution of the population on the three states coupled by two simultaneous few-cycle laser pulses with the same envelope but different CEPs. The parameters are the same as Fig. 5 but for $\omega_p = \omega_c = (1/3)\omega_{31} = (1/3)\omega_{32}$ corresponding to large detuning.

Then, let us discuss how the detuning impacts the time evolution of the population in our system. The case of large detuning for two simultaneous ultrashort pulses with different CEPs is illustrated in Fig. 7. It is assumed that all the carrier frequencies of the pump and Stokes field are equal to one third of the transition frequency ω_{31}. It is found that in some conditions the time evolution of the population coincides with the partial properties of an adiabatic process [see Figs. 7(a) and 7(c)]. This is induced by that the intermediate state $|3\rangle$ can be adiabatically eliminated for large detuning and the three-level system can be reduced as a two-level system. Based on these numerical results, we can see that the few-cycle pulses can be used to perform the population adiabatically in the case of large detuning. We simulate the model in which the laser field E_c is canceled and find that the effect of CEP introduced by the single pulse E_p is too week to be observed. Comparing Fig. 7(a) with Fig. 7(d), we can see that the CEP-dependent phenomenon is significant because of the quantum interference in the three-level system driven by two ultrashort pulses. This point is very useful for measuring the carrier envelope phase for few-cycle pulses without ionization.

The above cases considered the system are driven by an intuitive sequence of pulses and two simultaneous pulses. In the following, the case of the interaction between two sequential pulses with different delays will be studied. The impact of delay ΔT on the population dynamics is depicted in Figs. 8 and 9. Here, the pulse durations are different between these two figures and the shorter pulses are applied in Fig. 8. It should be noted that the pulse area is equal to π here. As in the former simulations, the CEPs also significantly affect the population distribution when the pulse duration is ultrashort. The reason is the same as that the level crossing occurs at different moments via Landau–Zener tunneling. The case of two longer pulses as inputs is illustrated in Fig. 9. We can see that the population is totally transferred to the target state $|2\rangle$ with the time delay increasing. The error rates of population transfer to the target state are about 3% and 0.5% in Figs. 8(d) and 9(d), respectively. However, the transfer process is non-adiabatic. For a large delay, the overlap of the sequential pulses is very small, which means that the sequential pulse can be considered as two separated pulses. In this case, the process of population transfer can be described as the following: the first pulse E_p transfers the population from state $|1\rangle$ to $|3\rangle$ via π pulse Ω_p; then the second pulse E_c transfers the population from state $|3\rangle$ to $|2\rangle$ via π pulse Ω_c. These points are clearly depicted in Fig. 8(d): when $T \simeq 1.3\times 2\pi/\omega_{31}$, $C_1 = C_2 = 0$, and $C_3 = 1$, which indicates that the population transfer from $|1\rangle$ to $|3\rangle$; when $T \simeq 4.5\times 2\pi/\omega_{31}$, $C_1 = C_3 = 0$ and $C_2 = 1$, which implies that the population transfers from $|3\rangle$ to $|2\rangle$. Hereto, we numerically investigate the population dynamics of Λ-type system in the presence of two few-cycle pulses and find that ultrashort pulses can be used to operate the population transfer. At the same time, our study shows that the quantum interference between two pulses can enlarge the signal of the CEP-dependent effect, which is helpful for estimating the value of CEP.

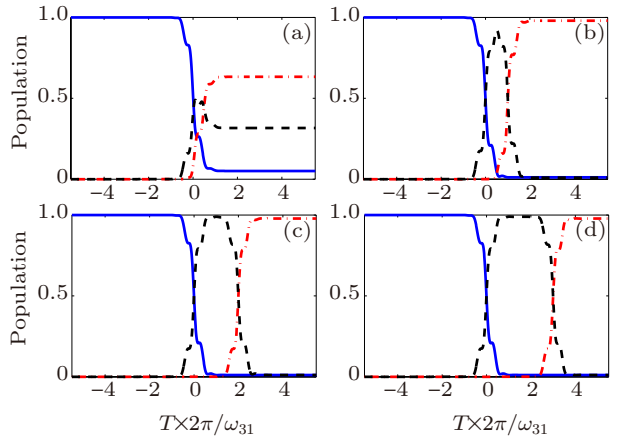


Fig. 8. (color online) Time evolution of the population on the three states coupled by two sequence few-cycle laser pulses with the same envelope but different delays. The solid, dash-dot, and dashed lines depict the population dynamics of states $|1\rangle$, $|2\rangle$, and $|3\rangle$, respectively. (a) $\Delta T = 0$; (b) $\Delta T = \pi/\omega_{31}$; (c) $\Delta T = 2\pi/\omega_{31}$; (d) $\Delta T = 3\pi/\omega_{31}$. Other parameters are: $\phi_p = 0$, $\phi_c = 0.5\pi$, $\omega_p = \omega_c = \omega_{31} = \omega_{32}$, $N_{p,c} = 1$, and $x_{31,32}E^0_{p,c}/\hbar = 0.4697\omega_{31}$ corresponding to π pulses.

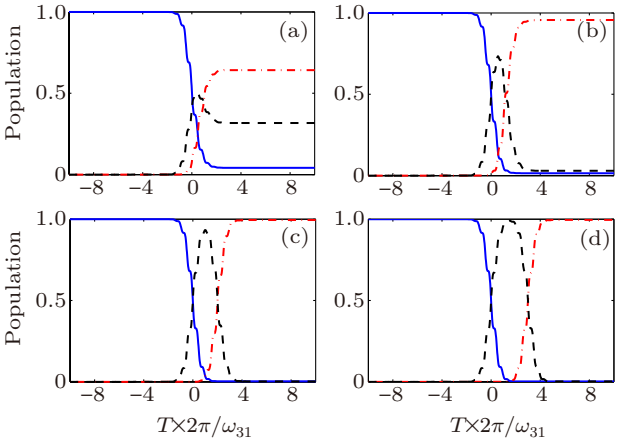


Fig. 9. (color online) Time evolution of the population on the three states coupled by two sequence few-cycle laser pulses with the same envelope but different delays. (a) $\Delta T = 0$; (b) $\Delta T = 2\pi/\omega_{31}$; (c) $\Delta T = 4\pi/\omega_{31}$; (d) $\Delta T = 6\pi/\omega_{31}$. The parameters are the same as these in Fig. 7 but for $N_{p,c} = 2$, $\phi_p = \phi_c = 0$, $x_{31,32}E^0_{p,c}/\hbar = 0.2349\omega_{31}$ corresponding to π pusles.

3. Approximate solution

In order to investigate the population dynamics discussed in Fig. 4, we will exhibit some theoretical analysis in this section. In normal conditions, the amplitudes of the few-cycle pulse are very strong. In other words, the laser field is varying fastly in a short time scale. The Landau–Zenel tunneling will occur in such process, as discussed before. It is difficult to obtain the analytic results. With the help of Magnus expansion,[53,54] the solution to the differential equations (1) in terms of the matrix $\boldsymbol{H}$ can be given as $\boldsymbol{C}(t) = \exp[\boldsymbol{M}]\boldsymbol{C}(t_0) = \exp[\boldsymbol{M}_1(t) + \boldsymbol{M}_2(t) + \cdots]\boldsymbol{C}(t_0)$ where $\boldsymbol{M}_1 = -\mathrm{i}\int_{t_0}^{t}\boldsymbol{H}(t_1)\mathrm{d}t_1$, $\boldsymbol{M}_2 = -(1/2)\int_{t_0}^{t}\int_{t_0}^{t_1}[\boldsymbol{H}(t_1), \boldsymbol{H}(t_2)]\mathrm{d}t_1\,\mathrm{d}t_2$. If $\boldsymbol{M}_2$ is equal to zero (in another word, $\int_{t_0}^{t}\boldsymbol{H}(t_1)\mathrm{d}t_1$ and $\boldsymbol{H}(t)$ are commutation), the solution for $\boldsymbol{C}(t)$ can be simplified to $\boldsymbol{C}(t) = \exp[\boldsymbol{M}_1(t)]\boldsymbol{C}(t_0)$. If $\boldsymbol{M}_2 + \cdots$ is much smaller than $\boldsymbol{M}_1$, the solution for $\boldsymbol{C}(t)$ can be approximated to the same expression $\boldsymbol{C}(t) = \exp[\boldsymbol{M}_1(t)]\boldsymbol{C}(t_0)$. Assumed $a_0(t) = \int_{t_0}^{t}\cos(\omega_{31}t')\Omega_p\mathrm{d}t'$, $a_1(t) = \int_{t_0}^{t}\sin(\omega_{31}t')\Omega_p\mathrm{d}t'$, $b_0(t) = \int_{t_0}^{t}\cos(\omega_{31}t' - \omega_{21}t')\Omega_c\mathrm{d}t'$ and $b_1(t) = \int_{t_0}^{t}\sin(\omega_{31}t' - \omega_{21}t')\Omega_c\mathrm{d}t'$, then equation (1) can be written as

$$\boldsymbol{C}(t) = U(t,t_0)\boldsymbol{C}(t_0) = \exp\left[-\mathrm{i}\tilde{H}\right]\boldsymbol{C}(t_0)\,. \tag{2}$$

Here, the exponential factor $\tilde{H}$ in the evolution operator $U(t,t_0)$ has the following form

$$\tilde{H} = \begin{pmatrix} 0 & 0 & a_0 - \mathrm{i}a_1 \\ 0 & 0 & b_0 - \mathrm{i}b_1 \\ a_0 + ia_1 & b_0 + \mathrm{i}b_1 & 0 \end{pmatrix}. \tag{3}$$

In order to obtain the analytical solution for the probability amplitudes, the matrix $\tilde{H}$ always can be decomposed as $\tilde{H} = Q\Lambda Q^{-1}$, where Λ is a diagonal matrix formed from the eigenvalues of $\tilde{H}$, and the columns of Q are the corresponding eigenvectors of $\tilde{H}$. Then we can obtain

$$U(t,t_0) = \exp\left[-iQ\Lambda Q^{-1}\right] = Q\exp[-\mathrm{i}\Lambda]Q^{-1}. \tag{4}$$

Here

$$Q = \begin{pmatrix} -\dfrac{b_0 + \mathrm{i}b_1}{a_0 + ia_1} & \dfrac{a_0 + \mathrm{i}a_1}{\Theta} & \dfrac{a_0 - \mathrm{i}a_1}{\Theta} \\ 1 & \dfrac{-b_0 + \mathrm{i}b_1}{\Theta} & \dfrac{b_0 - \mathrm{i}b_1}{\Theta} \\ 0 & 1 & 1 \end{pmatrix},$$

and

$$\Lambda = \begin{pmatrix} 0 & 0 & 0 \\ 0 & -\Theta & 0 \\ 0 & 0 & \Theta \end{pmatrix},$$

with $\Theta = \sqrt{a_0^2 + a_1^2 + b_0^2 + b_1^2}$. Then, we can obtain the analytical solution. If all the population lies on the state $|1\rangle$ on the initial moment, we can obtain the corresponding solution

$$\boldsymbol{C}_1(t) = \frac{2(b_0^2 + b_1^2) + (a_0^2 + a_1^2)[\mathrm{e}^{\mathrm{i}\Theta} + \mathrm{e}^{-\mathrm{i}\Theta}]}{2\Theta^2}, \tag{5}$$

$$\boldsymbol{C}_2(t) = \frac{(a_0 + \mathrm{i}a_1)(b_0 - ib_1)\mathrm{e}^{-\mathrm{i}\Theta}[-1 + \mathrm{e}^{\mathrm{i}\Theta}]^2}{2\Theta^2}, \tag{6}$$

$$\boldsymbol{C}_3(t) = \frac{(-a_0 - \mathrm{i}a_1)[-\mathrm{e}^{-\mathrm{i}\Theta} + \mathrm{e}^{\mathrm{i}\Theta}]}{2\Theta}. \tag{7}$$

Here, we will show the reason for complete population transfer via $\sqrt{2}\pi$ pulses. For simplicity, it is assumed that the two ultrashort pulses have the same envelope and carrier frequency which is equal to the transition frequency ω_{31}. Then, the wave functions for Ω_p and Ω_c can be read as $\Omega_p = f(t)\cos(\omega_{31}t)$ and $\Omega_c = f(t)\cos(\omega_{31}t)$. It is reasonable to ignore the CEPs according to the results in Fig. 4. If the CEPs are taken into account, the CEP-dependent effect would be investigated. At the same time, we assumed that the state $|1\rangle$ and $|2\rangle$ is degenerated ($\omega_{21} = 0$). After a trivial calculation, we can obtain that $a_0 = b_0 = (1/2)\int_{t_0}^{\infty} f(t')\mathrm{d}t'$ and $a_1 = b_1 = 0$ when

$t \to \infty$. In this derivation, we have used the fast-varying approximation to neglect the terms with fast oscillation. For the simulation in Fig. 4, the area of the input pulse is $\sqrt{2}\pi$. Then, we have $a_0 = b_0 = \sqrt{2}\pi/2$, $a_1 = b_1 = 0$, and $\Theta = \pi$ after the pulses go through the system. Substituting these results into Eq. (5), the final population distribution after these two simultaneous pulses is $|C_1(\infty)|^2 = |C_3(\infty)|^2 = 0$ and $|C_2(\infty)|^2 = 1$. This agrees well with the results in Fig. 4. Hereto, we have explained the reason for that $\sqrt{2}\pi$ pulses can be used to transfer the population to the target state. It is obviously that the phase dependent effect can be observed. If the CEPs are included in the expression Ω_c and Ω_c, a_0, a_1, b_0, and b_1 become dependent on the values of the CEPs. The phase Θ depends on the the rotating term in the Hamiltonian, which will introduce some quantum interferences in the population evolution [see Eqs. (5)–(7)] The results derived from Eqs. (5)–(7) agree well with the numerical results when the the time delay ΔT is small.

4. Summary

In summary, we illustrated the ultrafast population transfer of a Λ-configuration level system driven by few-cycle laser pulses. Different parameters are investigated, such as different pulse amplitudes and durations, different time delays between the two ultrashort pulses, and on(off) resonance. Our results show that the population transfer of a three-level system can be completed in the femtosecond time scale in some conditions. At the same time, a remarkable carrier-envelope-phase-dependent effect was found and the corresponding signal could be efficiently enlarged owing to the quantum interference effect induced by the ultrashort pulses.

Appendix A

In order to illustrate that the four-level system shown in Fig. 1(b) can be simplified as a three-level system as depicted in Fig. 1(a). Here, we consider a symmetric coupled double quantum well consisting of two 5.09-nm-wide GaAs wells, separated by a 1.13-nm-wide pure AlAs central barrier, and sandwiched between $Al_{0.3}Ga_{0.7}As$ layers.[44] The energy scheme of the corresponding quantum system has four bound states, as shown in Fig. 1(b). When the bias voltage is zero and the temperature is 45 K, the energy of the interband transition is about 1620 meV (766.9 nm), the energy gaps of the two excited levels and two ground states are 11 meV and 1 meV, respectively.[44] The dipole matrix element of the corresponding electron–hole transitions is $eD = 3.5$ e·nm.[55] If the Rabi frequency of the applied ultrashort pulses is about 0.4 time of the transition frequency as the parameter taken in Section 2, we can obtain the corresponding electric field amplitude being $\sim 1.87 \times 10^8$ V/m. According to the formulae $E = 2.74 \times 10^3 \sqrt{I[\mathrm{W/cm^2}]}$ V/m, the peak intensity is $\sim 5.73 \times 10^9$ W/cm^2. In such an intensity regime, the ionization can be neglected. We can see that the energy gap of the ground states is far less than the transition frequency.

For simplification, the dipole moments were set as the following $x_{3'1} = x_{3''1} = x_{31}/\sqrt{2}$ and $x_{3'2} = x_{3''2} = x_{32}/\sqrt{2}$. Here, x_{31} and x_{32} are the parameters taken in our former simulation. The factor $1/\sqrt{2}$ guarantees that the state $|3'\rangle$ and $|3''\rangle$ can be served as two sublevel states of state $|3\rangle$. In order to compare with the results calculated by using the three-level system, it is assumed that the two ultrashort pulses resonantly interact with the transitions $|3', 3''\rangle \longleftrightarrow |1, 2\rangle$. We calculated the population evolution of the ground states $|1\rangle$ and $|2\rangle$ in different situations. The numerical results are shown in Fig. A1. The impact of different transition frequencies of $|3''\rangle \longleftrightarrow |1, 2\rangle$ on population distribution were studied. When $\omega_{41} = \omega_{31}$, the case corresponds to the population evolution depicted in Fig. 2(a). From Fig. A1, we could find that the population of state $|1\rangle$ can be effectively transferred to state $|2\rangle$ even when $|\omega_{3''1} - \omega_{3'1}| = 0.05\omega_{31}$. The error of the population transfer is about 5% (see Figs. A1(a) and A1(d)). In the case simulated here, the energy gap $0.05\omega_{31}$ is much larger than the one for the symmetric coupled double quantum well discussed earlier.

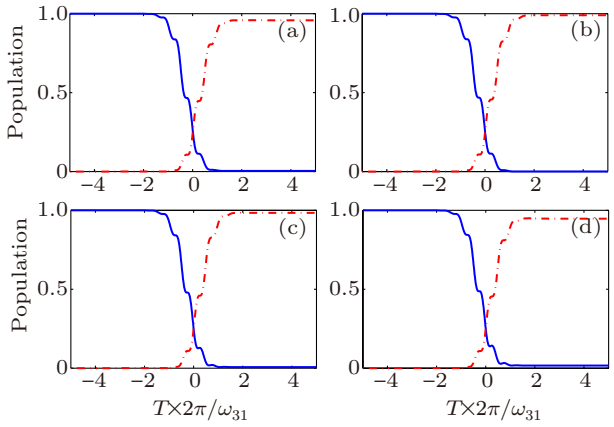


Fig. A1. (color online) Time evolution of the population on the states $|1\rangle$ and $|2\rangle$ coupled by an intuitive pair of few-cycle laser pulses with the same duration but for a four-level system. The solid line and dash-dotted line depict the population dynamics of states $|1\rangle$ and $|2\rangle$, respectively. (a) $\omega_{3''1} - \omega_{3'1} = -0.05\omega_{31}$; (b) $\omega_{3''1} - \omega_{3'1} = -0.025\omega_{31}$; (c) $\omega_{3''1} - \omega_{3'1} = 0.025\omega_{31}$; (d) $\omega_{3''1} - \omega_{3'1} = 0.05\omega_{31}$. Other parameters: $\phi_p = 0$, $\phi_c = 0$, $\omega_{3'1} = \omega_{31}$, $\omega_p = \omega_c = \omega_{31} = \omega_{32}$, $N_{p,c} = 2$, $\Delta T = 0$ and $x_{31,32}E^0_{p,c}/\hbar = 0.3321\omega_{31}$ corresponding to $\sqrt{2}\pi$ pulses. The curve of an ultrashort pulse where the CEP and cycle number are equal to 0 and 2, respectively.

Through calculations and comparisons, we find that the split energy levels can be idealized as a degenerated level in some cases. If $2\pi\hbar/\varepsilon \gg \tau$, such an idealization is valid. Based on this point, the three-level model we investigated former is reasonable and can be examined by experiments.

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