Gaussian and sinc-shaped few-cycle-pulse-driven ultrafast coherent population transfer in Λ-like atomic systems

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We report and propose a simple scheme to achieve efficient and fast coherent population transfer by utilizing either a nonlinearly chirped Gaussian-shaped few-cycle laser pulse or an unchirped sinc-shaped few-cycle laser pulse. The proposed scheme is shown to be fairly robust against the variation of the laser parameters such as temporal pulse width, chirp rates, carrier-envelope phases, and Rabi frequencies. We find that compared to the so-called stimulated Raman adiabatic passage technique, our scheme for complete population transfer with few-cycle Gaussian-shaped laser pulses requires less pulse area.

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I. INTRODUCTION

Coherent control of complete population transfer from an initial quantum state to a desired quantum state and creation of arbitrary coherence between two quantum states has been a major theme of atomic and molecular physics for quite some time now [1]. In particular, coherent control of population transfer is necessary for many well-known applications, including collision dynamics, atomic interferometry, spectroscopy, and optical control of chemical reactions, etc. Coherent population transfer (CPT) has found its relevance even in nuclear physics [2]. For robust and efficient controlling of population transfer between quantum states, many novel strategies have been proposed and exploited by several authors. Stimulated Raman adiabatic passage (STIRAP) [3–6], adiabatic rapid passage (ARP) [7], Raman chirped adiabatic passage (RCAP) [8–10], and temporal coherent control (TCC) [11,12], etc., are some of the well-known methods. Recently, the field of coherent control of atoms and molecules has received a tremendous boost owing to the recent progress in the generation of femtosecond and attosecond laser pulses and its possible future applications [13–18]. For example, quantum coherent control of physical and chemical processes and also of attosecond electronic dynamics by use of frequency- and amplitude-chirped few-cycle pulses is reported by some authors [19,20]. In the context of CPT, the use of these so-called ultrashort laser pulses has certain advantages, such as easy access to first electronic states of many molecules and an extremely fast transfer process which may be completed on a time scale much shorter than the typical time between collisions of atoms or molecules [21]. Cheng and Zhou [22] demonstrated numerically, without using the so-called rotating-wave approximation (RWA), the ultrafast population transfer in Λ-like three-state atomic systems with frequency-chirped few-cycle femtosecond laser pulses, though the scheme was not found to be robust against the variation of laser-pulse parameters. In passing, it should be mentioned that RWA may not hold when one deals with few-cycle-pulse-related phenomena and should work in the non-RWA regime [13,23–26]. Recently, Shapiro *et al.* [27,28] proposed a new

method, the so-called piecewise rapid adiabatic passage (PAP), for executing complete population transfer between quantum states in a piecewise manner using a series of femtosecond laser pulses. The proposed method is found to be robust against the variation in the absolute and relative intensities, durations, and time ordering of the pulses. More recently, Yang [24] exploited the STIRAP method to demonstrate the sufficiently robust and complete population transfer between the quantum states in Λ-like three-state atomic systems driven by few-cycle femtosecond laser pulses. However, for complete population transfer with STIRAP technique, the time-separated but partially overlapping pump and Stokes pulses should be applied in counterintuitive order, i.e., there should be complete control over the time lag between the pump and Stokes pulses. It is well known that the STIRAP technique is successfully implemented with continuous or narrow-band laser pulses. However, as discussed by many authors, when one needs to deal with few-cycle pulses or broadband pulses, this method may not be a suitable one [21,29,30]. Moreover, for complete population transfer between the quantum states, the STIRAP and rapid adiabatic passage are generally energetically expensive; for example, relative to a π−pulse technique [31,32]. On the other hand, sometimes the π−pulse technique is not robust against the variation of the laser-pulse parameters. In this work, we propose a relatively simple scheme, in which no time separation is required between the pump and Stokes pulses. Our method is energetically efficient and ultrafast. We show near complete CPT in the given Λ-like three-state atomic system which is driven by two simultaneously interacting nonlinearly chirped Gaussian-shaped few-cycle laser pulses or by two sinc-shaped unchirped few-cycle laser pulses, subject to the judicious choice of laser-pulse parameters. It may be mentioned that shaped pulses are also studied to enhance the transient population of excited states [26] and obtain total inversion of electronic state population in molecules [33]. Again, it may be noted that nonlinearly chirped laser pulses have been found suitable for many applications [34–38]. In this work, the phenomenon of coherent population transfer is investigated by numerically solving the appropriate density-matrix equations beyond the rotating-wave approximation. In Sec. II we present the optical Bloch equations that describe the interaction of the Λ system with the few-cycle laser pulses. Sec. III contains our simulated results and discussions followed by conclusions in Sec. IV.
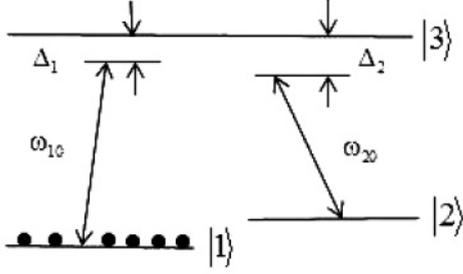
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FIG. 1. Schematic of Λ-like system with two acting few-cycle laser pulses.

II. THE MODEL

Our analysis is based on the scheme depicted in Fig. 1. We consider a Λ-like atomic system interacting with two few-cycle laser pulses. The electric field of the linearly polarized laser interacting between $|3\rangle$ and $|1\rangle$ is given by $\vec{E}_1 = \vec{E}_{10} f(t) \cos[\omega_{10} t + \delta_1(t)]$, where $\vec{E}_{10}$, $f(t)$, and ω_{10} are, respectively, the amplitude, the field envelope, and the carrier frequency of the pulse. Exactly analogous expression for the linearly polarized laser pulse interacting between $|3\rangle$ and $|2\rangle$ is given by $\vec{E}_2 = \vec{E}_{20} f(t) \cos[\omega_{20} t + \delta_2(t)]$. For the Gaussian-shaped few-cycle laser fields, $f(t) = \exp[-(t/\tau)^2]$, $\delta_1(t) = \chi_1 t^3$, and $\delta_2(t) = \chi_2 t^3$. Here χ_1 and χ_2 are the respective chirp rates of the Gaussian pulses. On the other hand, for the sinc-shaped few-cycle pulses, $f(t) = \sin(t/\tau)/(t/\tau)$ and $\delta_1 = \phi_1$ and $\delta_2 = \phi_2$, where ϕ_1 and ϕ_2 are the respective carrier-envelope phases. τ_p is the temporal width of the laser pulses. For the Gaussian pulse, $\tau_p = 1.177\tau$, while $\tau_p = 2.783\tau$ for the sinc pulse.

In the given scheme, we assume that only $|3\rangle \rightarrow |1\rangle$ and $|3\rangle \rightarrow |2\rangle$ transitions are dipole allowed, while $|2\rangle \rightarrow |1\rangle$ transitions are forbidden. The Hamiltonian of the system is given by $\widehat{H} = \widehat{H}_0 + \widehat{H}_{\text{int}}$, where $\widehat{H}_0 = \hbar\omega_1|1\rangle\langle1| + \hbar\omega_2|2\rangle\langle2| + \hbar\omega_3|3\rangle\langle3|$ and $\widehat{H}_{\text{int}} = -\vec{\mu} \cdot \vec{E} = -\hbar\Omega_{31}(t)|3\rangle\langle1| - \hbar\Omega_{32}(t)|3\rangle\langle2| + \text{H.c.}$

Here $\Omega_{31}(t) = \mu_{31}\vec{E}_1(t)/\hbar$ and $\Omega_{32}(t) = \mu_{32}\vec{E}_2(t)/\hbar$ are the time-dependent Rabi frequencies for the transition with electric dipole moment μ_{31} and μ_{32}, respectively. The Bloch equations, without invoking the so-called rotating-wave approximation, describing the temporal evolution of the density matrix elements, are

$$
\begin{aligned}
\dot{\rho}_{31} &= -i\omega_{31}\rho_{31} + i\Omega_{32}(t)\rho_{21} - i\Omega_{31}(t)(\rho_{33} - \rho_{11}), \\
\dot{\rho}_{32} &= -i\omega_{32}\rho_{32} + i\Omega_{31}(t)\rho_{12} - i\Omega_{32}(t)(\rho_{33} - \rho_{11}), \\
\dot{\rho}_{21} &= -i\omega_{21}\rho_{21} + i\Omega_{32}(t)\rho_{31} - i\Omega_{31}(t)\rho_{23}, \\
\dot{\rho}_{11} &= i\Omega_{31}(t)(\rho_{31} - \rho_{13}), \\
\dot{\rho}_{22} &= i\Omega_{32}(t)(\rho_{32} - \rho_{23}), \\
\dot{\rho}_{33} &= i\Omega_{31}(t)(\rho_{13} - \rho_{31}) + i\Omega_{32}(t)(\rho_{23} - \rho_{32}).
\end{aligned}
\tag{1}
$$

Here $\omega_{ij} = \omega_i - \omega_j$. It may be noted that $\rho_{ij} = \rho_{ji}^*$.

III. RESULTS AND DISCUSSIONS

We solve Eq. (1) numerically using a standard fourth-order Runge-Kutta method. We assume that initially all the atoms are in the ground state $|1\rangle$. We use the following typical
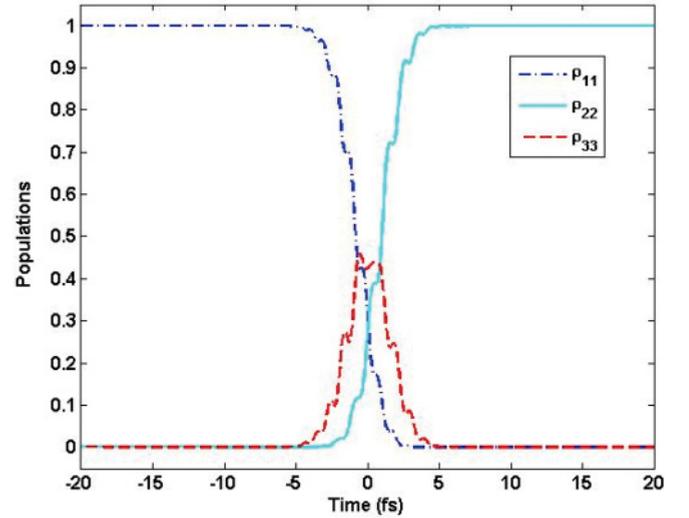

FIG. 2. (Color online) Temporal evolution of populations with the nonlinearly chirped Gaussianshaped few-cycle pulse.

parameters: $\omega_{31} = \omega_{10} = 3.0$ rad/fs, $\omega_{21} = 0.4$ rad/fs, $\omega_{32} = \omega_{20} = 2.6$ rad/fs, $\Omega_{31} = 0.76$ rad/fs, $\Omega_{32} = 0.79$ rad/fs, $\chi_1 = \chi_2 = 0.016$ fs^{-3}, and $\phi_1 = \phi_2 = 0$. The temporal pulse width is taken to be $\tau_p = 4.70$ and 5.06 fs, respectively, for the Gaussian and the sinc pulses. For our chosen parameters, the pulse areas are too small for the so-called adiabatic condition to be fulfilled. It is worthwhile to note that in the usual adiabatic passage scheme for population transfer between the initial state and the final one, the adiabatic condition can be written as $\Omega\tau \gg \pi$, where $\Omega = \Omega_{31}(0) = \Omega_{32}(0)$ is the maximal Rabi frequency [39]. In practical applications the pulse area should exceed 10π, i.e., $\Omega\tau > 10\pi$ to provide efficient population transfer via the adiabatic passage scheme [40]. We find that for complete population transfer, with our proposed scheme, the total temporal area of the Gaussian pulses is calculated to be 3.49π, while it is 13.53π for the STIRAP scheme with same laser-pulse parameters. Figures 2 and 3 depict the respective
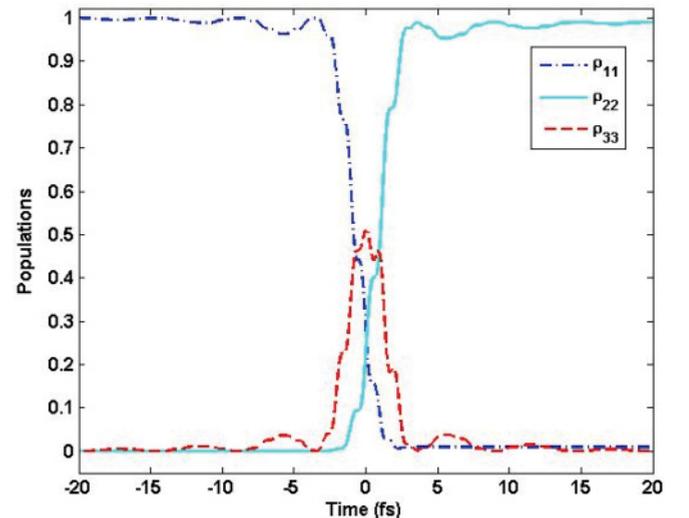

FIG. 3. (Color online) Temporal dynamics of populations with the sinc-shaped few-cycle pulse.

temporal evolution of the populations ρ_{11}, ρ_{22}, and ρ_{33} when chirped Gaussian pulses and unchirped sinc pulses are used.

It is clear from Fig. 2 that one can obtain complete population transfer (99.94%) from the ground state $|1\rangle$ to the state $|2\rangle$ using two nonlinearly chirped Gaussian-shaped laser pulses. On the other hand, as evident from Fig. 3, near complete population transfer (99.05%) from $|1\rangle$ to $|2\rangle$ is possible due to the simultaneous interaction of two sinc-shaped pulses with the three-level atomic system.

These results could be explained on the basis of the so-called stimulated emission pumping (SEP) [31]. In SEP with continuous laser or short laser pulses, all the relaxation processes in atomic systems take place on the time scale shorter than the interaction time. Hence the maximum amount of population transfer between the quantum states is restricted by the spontaneous emission. For example, one can achieve maximum 30% population transfer in Λ-like three-level atomic systems with SEP technique [31]. However, in our scheme, we have shown that almost complete population transfer in Λ-like atomic systems is possible owing to the use of few-cycle pulses where interaction takes on a time scale shorter than that of the relaxation processes. Nonadiabatic consequences on the temporal evolution of the populations could be observed from Figs. 2 and 3. Unlike the adiabatic passage techniques, population in quantum state $|3\rangle$ during the intermediate time is approaching a large value ($\rho_{33} = 45\%$ for Gaussian pulse and $\rho_{33} = 46\%$ for sinc pulse) as the adiabatic criteria is not fulfilled for the chosen laser-pulse areas. However, finally the quantum state $|2\rangle$ receives almost all the populations in both cases. The non-RWA effects on the temporal evolution of the populations could also be observed from Figs. 2 and 3. Some authors have pointed out that when the few-cycle laser pulses are considered, the time-derivative-driven nonlinearities will have a significant impact on the interaction of the laser pulses with the atomic medium which may lead to strong oscillation features during the evolution of the populations [41,42]. These features are not present in the RWA solutions. Now, in order to have some insight into or understanding of why the use of Gaussian-shaped or sinc-shaped few-cycle-pulse results in almost similar behavior with regard to population transfer, in Fig. 4 we plot the temporal evolution of both the pulses.

Figure 4 reveals that the nonlinearly chirped Gaussian and the unchirped sinc-shaped pulses are equivalent to each other, particularly in the temporal range from -2 to $+2$ fs. For other temporal ranges from -6 to -2 fs and -2 to $+6$ fs, the amplitude of the nonlinearly chirped Gaussian pulse is slightly greater than that of the unchirped sinc-shaped pulse. Also, the carrier oscillation frequency of the Gaussian-shaped few-cycle pulse is slightly larger, owing to the nonlinear chirp, than that of the unchirped sinc-shaped few-cycle pulse in that temporal range. This might be the reason behind the almost similar nature of interaction of the pulses with the atomic system. Hence, we may conclude that, subject to the chosen parameters, the nonlinearly chirped Gaussian and the unchirped sinc-shaped few-cycle pulses exhibit almost identical behavior. It may be observed from Fig. 4 that one-($\Delta_1 = \omega_{31} - \omega_{10} = \Delta_2 = \omega_{32} - \omega_{20} = 0$) and two-photon ($\Delta = \Delta_1 - \Delta_2 = 0$) resonance conditions are fulfilled for unchirped sinc-shaped laser pulses. However, for the nonlinearly chirped Gaussian laser pulse, one photon resonance condition is partially
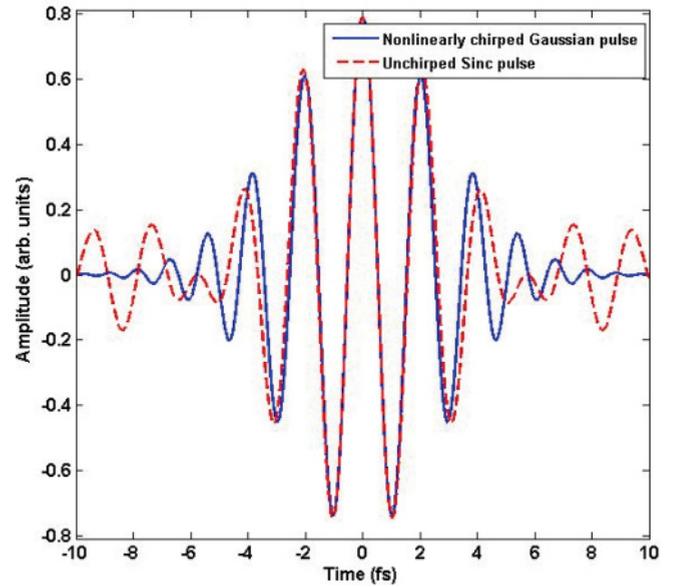

FIG. 4. (Color online) Temporal evolution of nonlinearly chirped Gaussian and unchirped sinc-shaped pulses.

fulfilled during the intermediate time of interaction while the two-photon resonance condition is fulfilled during the whole interaction. It is important to verify the robustness of the scheme against the variation of the chirp rate and the temporal pulse width of the Gaussian-shaped few-cycle pulse. So, in Fig. 5 we present the simulation result for the variation of the final population transfer to the state $|2\rangle$, i.e., $\rho_{22}(\infty)$ with τ_p, while in Fig. 6 we check the robustness of the scheme against chirp rates for the nonlinearly chirped Gaussian-shaped few-cycle pulses. It can be seen from Fig. 5(a) that the final population transfer $\rho_{22}(\infty)$ to the quantum state $|2\rangle$ is sufficiently robust against the variation (4–6 fs) of temporal width of Gaussian pulse, while Fig. 5(b) shows that the final population transfer $\rho_{22}(\infty)$ to quantum state $|2\rangle$ is robust against the small variation (4.94–5.17 fs) of temporal width of the sinc pulse.
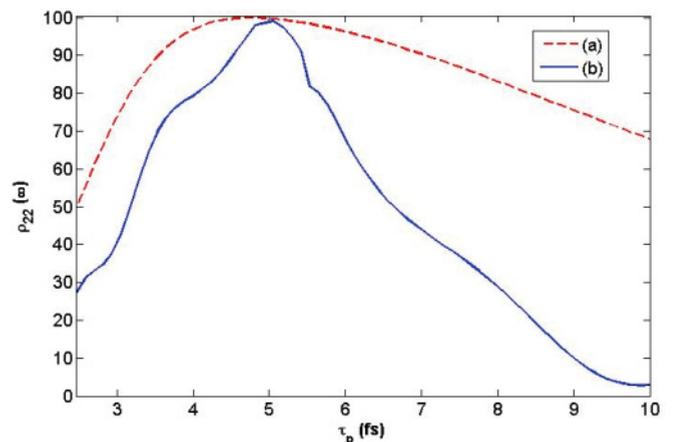

FIG. 5. (Color online) Final population to quantum state $|2\rangle$ as a function of (a) temporal pulse width τ_p of the nonlinearly chirped Gaussian pulse and (b) temporal pulse width τ_p of the sinc pulse. Here all the other parameters are kept constant.
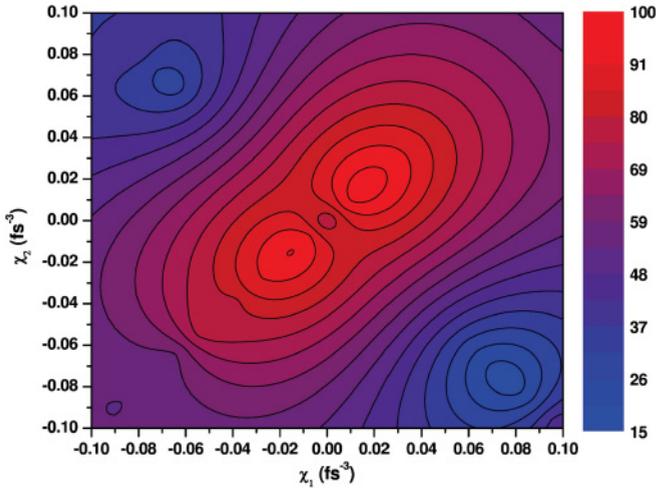
FIG. 6. (Color online) Final population transfer $\rho_{22}(\infty)$ to quantum state $|2\rangle$ as a function of chirp rates χ_1 and χ_2, and other parameters are the same as those in Fig. 2.

It could be observed from Fig. 6 that the final population transfer $\rho_{22}(\infty)$ is sufficiently robust against the variation of the chirp rates χ_1 and χ_2. It can be seen that the final population transfer $\rho_{22}(\infty)$ to the quantum state $|2\rangle$ is robust against the small variation (0.012–0.020 fs^{-3}) of the chirp rates χ_1 and χ_2.

In Fig. 7 we plot the variation of the final population $\rho_{22}(\infty)$ with the carrier-envelope phases to check the robustness of the scheme for sinc-shaped few-cycle pulses. We find that, from Fig. 7, the final population transfer $\rho_{22}(\infty)$ is highly robust against the variation of the carrier-envelope phases ϕ_1 and ϕ_2 of the sinc-shaped few-cycle pulses.

Finally, we test the robustness of our proposed scheme against variation of the Rabi frequencies of the few-cycle laser pulses considered in this work. In Fig. 8, we depict the contour map of the final population to the quantum state $|2\rangle$, i.e., $\rho_{22}(\infty)$, against Rabi frequencies Ω_{31} and Ω_{32} of the nonlinearly chirped Gaussian-shaped few-cycle

FIG. 8. (Color online) Contour maps of the final population (in %) transfer for varying Rabi frequencies Ω_{31} and Ω_{32} of nonlinearly chirped Gaussian pulse, and other parameters are the same as those in Fig. 2.

pulse. A careful inspection of Fig. 8 reveals that the final population transfer is fairly robust against the variation of the Rabi frequencies Ω_{31} and Ω_{32} in the range 0.70–0.92 rad/fs, which amounts to more than 95% population. Population in the range 88%–90% could be possible for variation of the Rabi frequencies in the range 2.30–2.40 rad/fs. A similar contour map for the sinc-shaped few-cycle pulse is depicted in Fig. 9. The population transfer to the quantum state $|2\rangle$ exhibits sufficient robustness with the small variation of Rabi frequencies. In fact one can obtain $> 95\%$ population transfer for the variation of Ω_{31} and Ω_{32} in the range 0.70–0.85 rad/fs.

It is worthwhile to mention that we have tested our proposed scheme for atomic systems such as indium, thallium, and gallium. We observe near complete population transfer, with
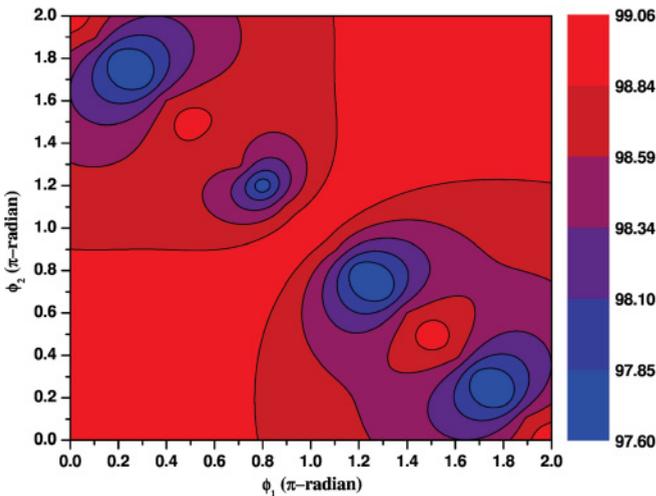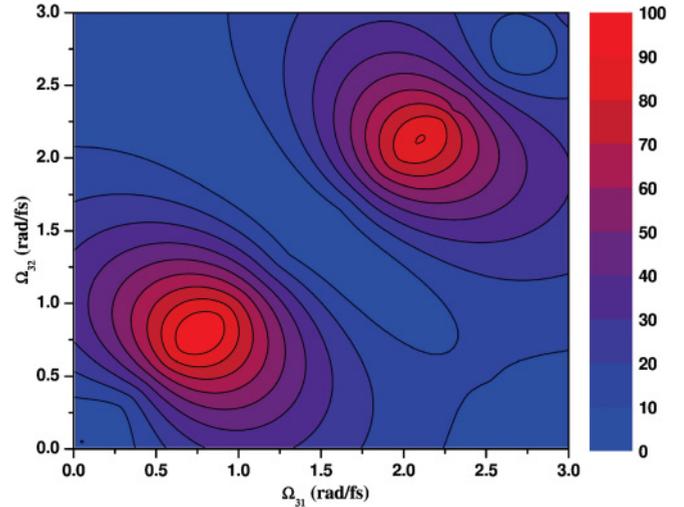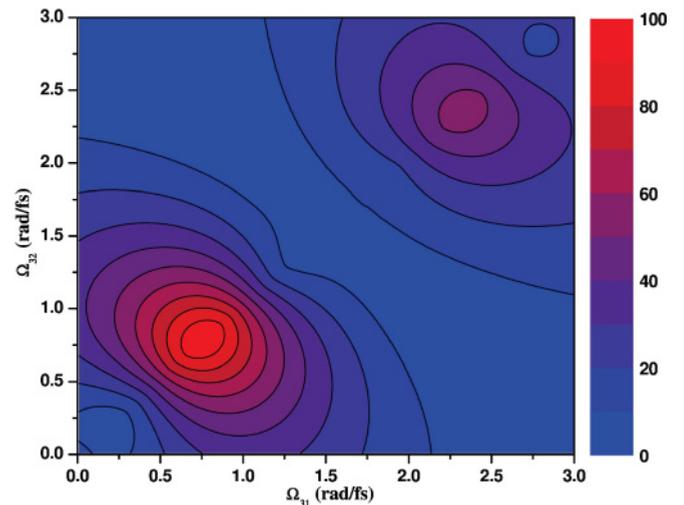

FIG. 7. (Color online) Final population transfer $\rho_{22}(\infty)$ to quantum state $|2\rangle$ as a function of (a) carrier-envelope phases ϕ_1 and ϕ_2 in the unit of π radian, and other parameters are the same as those in Fig. 3.

FIG. 9. (Color online) Contour maps of the final population (in %) transfer for varying Rabi frequencies Ω_{31} and Ω_{32} of unchirped sinc pulse, and other parameters are the same as those in Fig. 3.

appropriate choice of simulation parameters, in these atomic systems with both Gaussian and sinc-shaped few-cycle pulses. Hence the model proposed by us may be tested experimentally in atomic systems such as indium, thallium, and gallium. In passing, it may be noted that indium, thallium and gallium have been studied extensively both theoretically and experimentally in the context of laser cooling of atoms [43–46].

IV. CONCLUSIONS

We have demonstrated almost complete population transfer to the target quantum state with nonlinearly chirped Gaussian-shaped and unchirped sinc-shaped few-cycle laser pulses even when the adiabatic condition is not fulfilled. Population transfer with nonlinearly chirped Gaussian pulse is found to be sufficiently robust against the variation of temporal pulse width, Rabi frequencies, and the chirp rates. However, the population transfer with unchirped sinc-shaped pulse is found to be highly robust against the variation of carrier-envelope phase and fairly robust against the variation of the temporal pulse width and Rabi frequencies. We find that compared to the so-called STIRAP technique, our scheme for complete population transfer with few-cycle Gaussian-shaped laser pulses require less pulse area. Hence, nonlinearly chirped Gaussian-shaped few-cycle laser pulses or the sinc-shaped few-cycle laser pulses even with highly fluctuating carrier-envelope phase, may be employed for an efficient and ultrafast coherent population transfer with judicious choice of laser-pulse parameters.

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