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Ultrafast coherent population transfer driven by two few-cycle laser pulses

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Abstract. We investigate ultrafast coherent population transfer driven by few-cycle pump and Stokes laser pulses in the Λ-type three-level system with the stimulated Raman adiabatic passage technique beyond the rotating-wave approximation. In contrast to the case with the rotating wave approximation, the most efficient population transfer may be realized without the satisfaction of the one-photon resonances or two-photon resonance and the transfer efficiency depends critically on the Rabi frequencies and initial optical phases of the two laser fields when the peak Rabi frequencies are much larger than the respective transition frequencies. Moreover, complete and robust population transfer can still be obtained with the variations of the Rabi frequencies, pulse durations, and one-photon or two-photon detuning in a moderate range, though a considerable transient population may reside in the excited state. These abnormal behaviors result from the counterrotating terms, which are not taken into account in the traditional rotating wave approximation.

The stimulated Raman adiabatic passage (STIRAP) has proven to be an efficient and robust way for selective and complete coherent population transfer between two discrete atomic or molecular states [1–6]. It is well known that, under the traditional rotating wave approximation (RWA), three conditions should be satisfied in order to realize perfect population transfer in the simplest Λ-type or ladder-type three-level system with the STIRAP technique. Firstly, the time-separated but partially-overlapping pump and Stokes pulses should be applied in the counterintuitive order (i.e., the Stokes pulse precedes the pump pulse); secondly, the two-photon resonance between the initially populated state 1 and target state 3 should be maintained (while the intermediate state 2 may be off resonance by a certain detuning); thirdly, the time evolution of the atomic or molecular system should be adiabatic, that is, the pulse area should be large enough (normally larger than 10) [1–3]. However, for the case of few-cycle laser pulses or the Rabi frequencies of the laser fields comparable to or larger than the respective transition frequencies or the difference of them, the RWA may fail to work and the counterrotating terms should be taken into account. The effects of the counterrotating terms on coherent population transfer have been extensively studied. Kaluza and Muckerman [7], Casperson [8], and Genkin [9] investigated the interaction of a two-level system with the few-cycle laser pulse without the RWA. It was pointed out in references [10,11] that coherent population trapping beyond the RWA can still exist under certain conditions in the Λ-type three-level system. Cheng and Zhou [12] considered ultrafast population transfer in the Λ-type three-level system driven by a chirped few-cycle pulse, and demonstrated that the maximal population transfer depends strongly on the chirp rate and intensity of the pulse. It was shown that complete population transfer without the RWA can be achieved as well under certain conditions in the Λ-type three-level system with the STIRAP technique when the Rabi frequencies of the laser fields are comparable to or larger than the respective transition frequencies [13,14] or the difference of them [15,16].

In the previous studies [13–16] on the coherent population transfer in the Λ-type three-level system with the STIRAP technique beyond the RWA, the one-photon resonances or two-photon resonance is often assumed, which is generally thought to be one necessary condition for efficient population transfer. Nevertheless, in this paper, we demonstrate that the most efficient population transfer may be realized without the satisfaction of the one-photon resonances or two-photon resonance in the Λ-type three-level system driven by few-cycle pump and Stokes laser pulses with the STIRAP technique. Moreover, in references [13–16], the pulse durations are much longer than the optical periods of the laser fields (i.e., the multi-cycle pulses are used), where the initial optical phases of the laser fields are not taken into account. In addition, as is well known, the transfer efficiency exhibits robustness to the variations of the Rabi frequencies and pulse durations

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$$H = \hbar \begin{pmatrix} 0 & \Omega_p^*(t)\cos(\omega_p t + \varphi_p) & 0 \\ \Omega_p(t)\cos(\omega_p t + \varphi_p) & \omega_{21} & \Omega_S(t)\cos(\omega_S t + \varphi_S) \\ 0 & \Omega_S^*(t)\cos(\omega_S t + \varphi_S) & \omega_{31} \end{pmatrix} \quad (1)$$

$$H^{'} = \hbar \begin{pmatrix} 0 & \frac{\Omega_p^*(t)}{2}\left(1 + e^{-j2\left(\omega_p t + \varphi_p\right)}\right) & 0 \\ \frac{\Omega_p(t)}{2}\left(1 + e^{j2\left(\omega_p t + \varphi_p\right)}\right) & -\Delta_1 & \frac{\Omega_S(t)}{2}\left(1 + e^{j2(\omega_S t + \varphi_S)}\right) \\ 0 & \frac{\Omega_S^*(t)}{2}\left(1 + e^{-j2(\omega_S t + \varphi_S)}\right) & \Delta_2 - \Delta_1 \end{pmatrix} \quad (2)$$

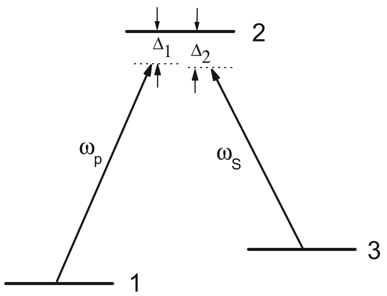

Fig. 1. The Λ-type three-level system. Levels 1 and 2 is coupled by the pump pulse 1 and levels 3 and 2 by the Stokes pulse.

in the adiabatic limit under the RWA. However, here we show that the population transfer efficiency depends critically on the initial optical phases and Rabi frequencies of the laser fields when the Rabi frequencies are much larger than the respective transition frequencies.

The considered Λ-type three-level system, as shown in Figure 1, interacts with two time-separated but partially-overlapping laser pulses, where level 1 and upper level 2 is coupled by the pump pulse and levels 3 and 2 is coupled by the Stokes pulse. We assume the electric field $E_S(t)$ of the Stokes pulse can be written as $E_S(t) = E_{0s} f(t)\cos(\omega_S t + \varphi_S)$, where ω_S is the carrier frequency, $f(t)$ is the electric-field amplitude envelope with the peak value of E_{0S}, and φ_S is the initial optical phase of the Stokes field. For simplicity, we assume the pump and Stokes pulses have the same envelope functions, and the pump pulse is delayed from the Stokes pulse by τ, so the electric field $E_p(t)$ of the pump pulse can be expressed as $E_p(t) = E_{0p} f(t - \tau)\cos(\omega_p t + \varphi_p)$. The Rabi frequencies of the pump and Stokes pulses are assumed to be Gaussian with the amplitude envelopes of the form $\Omega_S(t) = \Omega_{0S}\exp(-t^2/T^2)$ and $\Omega_p(t) = \Omega_{0p}\exp[-(t-\tau)^2/T^2]$, respectively, where T is the pulse duration and $\Omega_{0p(S)} = \mu_{1(2)} E_{0p(S)}/\hbar$ is the peak value of the Rabi frequency of the pump (Stokes) pulse with $\mu_1(\mu_2)$ being the dipole moment for the transition $1{-}2$ $(3{-}2)$. The detunings of the pump and Stokes lasers from the resonant transitions $1{-}2$ and $3{-}2$ are given by $\Delta_1 = \omega_p - \omega_{21}$ and $\Delta_2 = \omega_S - \omega_{23}$, respectively, where $\omega_{ij} (i \neq j)$ is the resonant frequency between levels i and j.

The time-dependent Hamiltonian of the matter-field system can be written as

see equation (1) above.

As is commonly done, by introducing the three-state rotating wave transformation,

$$\Psi(t) = c_1(t)\left|1\right\rangle + c_2(t)\exp\left[-j\left(\omega_p t + \varphi_p\right)\right]\left|2\right\rangle + c_3(t)\exp\left[-j\left(\omega_p t - \omega_S t + \varphi_p - \varphi_S\right)\right]\left|3\right\rangle,$$

We can get the transformed Hamiltonian $H^{'}$,

see equation (2) above.

In the above Hamiltonian, we do not apply the RWA. Obviously, as compared to the traditional Hamiltonian with the RWA [1–6], the inclusion of both the rotating and counterrotating terms in the Hamiltonian in equation (2) is equivalent to replacing the Rabi frequencies $\Omega_p(t)$ and $\Omega_S(t)$ in the usual Hamiltonian under the RWA by $\Omega_p(t)(1 + e^{j2(\omega_p t + \varphi_p)})$ and $\Omega_S(t)(1 + e^{j2(\omega_S t + \varphi_S)})$, respectively. As analyzed in reference [14], the consideration of the counterrotating terms corresponds to the introduction of amplitude and phase modulations to the Rabi frequencies of the two laser pulses. It is quite evident that when the Rabi frequencies $\Omega_p(t)$ and $\Omega_S(t)$ are far smaller than the corresponding transition frequencies, the counterrotating terms can be neglected and the Hamiltonian $H^{'}$ reduces to the usual description in the RWA. In the following, we consider the situation that when the Rabi frequencies $\Omega_p(t)$ and $\Omega_S(t)$ are comparable to or much larger than the respective transition frequencies, how the counterrotating terms affect coherent population transfer in the Λ-type three-level system driven by few-cycle pump and Stokes laser pulses with the STIRAP technique. We resolve the time-dependent Schrödinger equation with the fourth-order Runge-Kutta integrator. In what follows, the population is assumed to be initially in level 1, and the time and frequency are scaled with fs and fs^{-1}, respectively.

Figure 2 shows the time evolution of the populations in the three states 1, 2, and 3 with the two laser fields tuned to resonance with the respective transitions with the time delay equal to pulse duration $\tau = T = 5$, initial optical phases $\varphi_S = \varphi_p = 0$, resonant transition frequencies

$\omega_{21} = \pi$ (the corresponding optical wavelength of 600 nm) and $\omega_{23} = 0.8\pi$, and Rabi frequencies $\Omega_{0S} = \Omega_{0p} = 1.2$ or $\Omega_{0S} = \Omega_{0p} = 5$ for the cases with and without the RWA. With these parameters of the pulses, the standard STIRAP condition [1–6] for adiabatic evolution is fulfilled. Note that, according to reference [17], the required Rabi frequencies nearly comparable to the transition frequencies for a 5 fs laser pulse can be experimentally obtained. In all figures, the time origin is chosen at the peak of the Stokes pulse. As seen in Figures 2a and 2c, when the pulse areas are about in the low limit of the adiabatic criterion (about equal to 10) [1–3], nearly complete population transfer can be realized with the counterintuitively-ordered and partially-overlapping few-cycle pump-Stokes pulses in the cases both with and without the RWA, though an appreciable transient population would reside in the excited state 2 due to the limited pulse areas. However, there exists a distinct difference between the two cases, that is, the curves of the populations during the two pulses excitation are very smooth under the RWA (see Fig. 2a), whereas fast oscillations of the populations appear in the case without the RWA (see Fig. 2c), which results from the counterrotating terms. The similar fast oscillations have also been observed in the two-level [8,9] and three-level systems [13–15] driven by few-cycle pulses. When the pulse areas are much larger (e.g., about equal to 44 in Figs. 2b and 2d), perfect population transfer can occur and nearly no transient population would enter the excited state 2 under the RWA (see Fig. 2b); nevertheless, beyond the RWA, the populations in the three states undergo rapid irregular variations during the two pulses excitation and the transfer efficiency is very low (the final populations in the states 1, 2, and 3 are about 69%, 23%, and 8%, respectively), as shown in Figure 2d. This clearly indicates the invalidity of the RWA for the few-cycle pulses excitation with much larger Rabi frequencies. Note that as the interaction time (about a few femtoseconds) is far smaller than the population decay time (normally about tens of ps to tens of ns), the population decay of the excited state 2 nearly has no effect on the transfer efficiency.

As is well known, under the RWA, the population transfer in the Λ-type three-level system with the STIRAP technique exhibits robustness to the variations of the Rabi frequencies and pulse durations of the laser fields in the adiabatic regime, and the larger the pulse areas, the more efficient the population transfer. In order to see whether such population transfer with robustness to the changes of Rabi frequencies and pulse durations can take place for the case beyond the RWA, we show in Figure 3b the contour plot of the population transfer efficiency as a function of the Rabi frequencies $\Omega = \Omega_{0p} = \Omega_{0S}$ and pulse durations T with the two laser fields tuned to resonance with the respective transitions, and the other parameters scaled with fs and fs^{-1} are the same as those in Figure 2. It is clear from Figure 3b that when the Rabi frequencies (or pulse durations) are comparable to or a little larger than the transition frequencies (or the optical periods of the laser fields), robust and nearly perfect population transfer can be obtained with the variations of the pulse durations and Rabi frequencies in a certain range. However, when the Rabi frequencies are much larger than the transition frequencies, the transfer efficiency does not have the robustness to the pulse areas which characterize the traditional STIRAP with the RWA, and exhibits a complicated pattern with respect to the Rabi frequencies and pulse durations. For comparison, we show in Figure 3a the contour plot of the population transfer efficiency as a function of

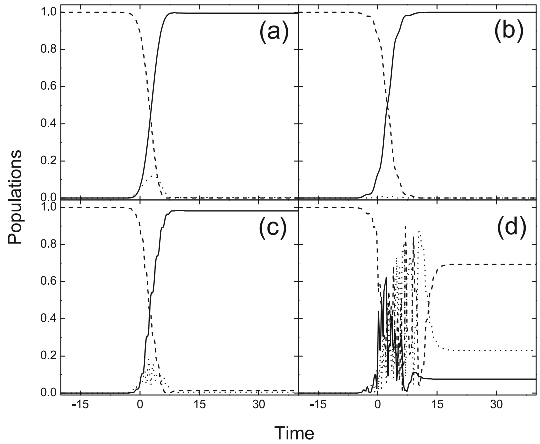

Fig. 2. The time evolution of the populations in the three states 1 (dashed line), 2 (dotted line), and 3 (solid line) with the two laser fields tuned to resonance with the corresponding transitions under the Rabi frequencies of $\Omega_{0S} = \Omega_{0p} = 1.2$ ((a) and (c)) and $\Omega_{0S} = \Omega_{0p} = 5$ ((b) and (d)) with $\tau = T = 5$, $\varphi_S = \varphi_p = 0$, $\omega_{21} = \pi$, and $\omega_{23} = 0.8\pi$ (in corresponding units of fs or fs^{-1}), for ((a) and (b)) with the RWA, and ((c) and (d)) without the RWA.

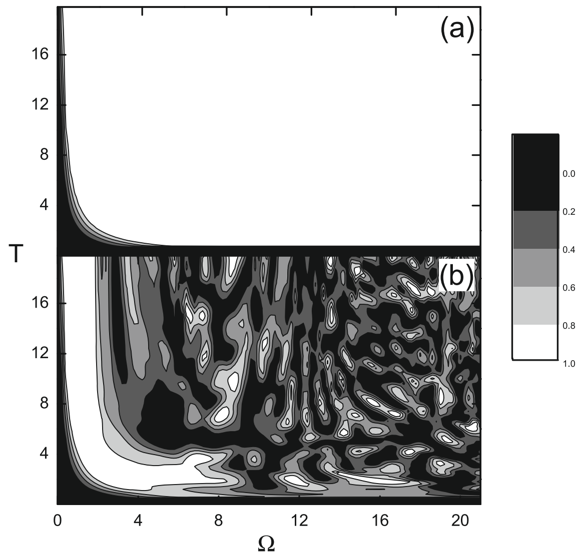

Fig. 3. The contour plots of the population transfer efficiencies as a function of the Rabi frequencies $\Omega = \Omega_{0p} = \Omega_{0S}$ and pulse durations T with the two laser fields tuned to resonance with the respective transitions, for (a) without the RWA, and (b) with the RWA, and the other parameters are the same as those in Figure 2.

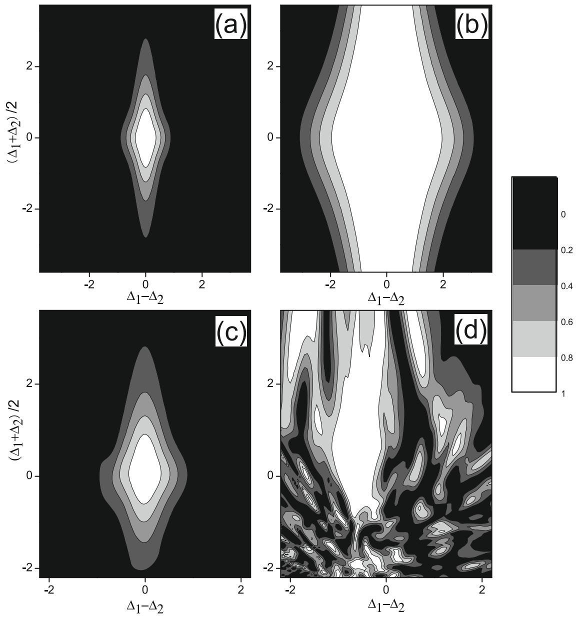

Fig. 4. The contour plots of the population transfer efficiencies as a function of the one-photon detuning ($\frac{\Delta_1+\Delta_2}{2}$) and two-photon detuning ($\Delta_1 - \Delta_2$) with the Rabi frequencies $\Omega_{0S} = \Omega_{0p} = 1.2$ ((a) and (c)), and $\Omega_{0S} = \Omega_{0p} = 5$ ((b) and (d)), for ((a) and (b)) with and ((c) and (d)) without the RWA, and the other parameters are the same as those in Figure 2.

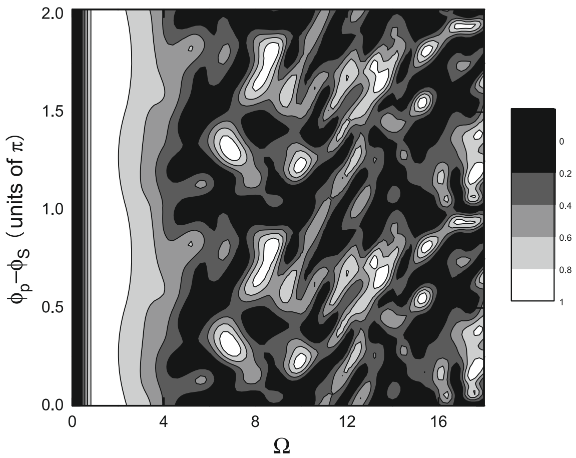

Fig. 5. The contour plot of the population transfer efficiency beyond the RWA as a function of the Rabi frequencies $\Omega = \Omega_{0p} = \Omega_{0S}$ and phase difference $\varphi = \varphi_p - \varphi_S$ of the two laser fields with the fields tuned to resonance with the respective transitions and $\varphi_S = 0$, and the other parameters are the same as those in Figure 2.

the Rabi frequencies and pulse durations under the RWA, where the large white region means the robustness of the transfer efficiency to the pulse durations and Rabi frequencies. The discrepancy further confirms the invalidity of the RWA for few-cycle pulses excitation with much larger Rabi frequencies. The intricate dependence of population transfer on the Rabi frequencies, which results from the counterrotating terms, can be well understood with the adiabatic Floquet theory [13–16]. As analyzed in references [13–16], when the Rabi frequencies are much larger than the respective transition frequencies, the nonadiabatic coupling between the Floquet states may occur due to the counterrotating terms, which would lead to diabatic evolution of the transfer state to the other Floquet states and subsequent complicated population transfer with respect to the Rabi frequencies.

It is also well known that in the conventional STIRAP with the RWA, one essential condition is that the two-photon resonance should be maintained, and the most efficient population transfer takes place when the two laser fields are tuned to resonance with their respective transitions. To see how the one-photon and two-photon detunings affect the population transfer for the case of few-cycle pulses excitation when the counterrotating terms are taken into account, we show in Figure 4 the contour plots of the population transfer efficiencies as a function of the one-photon detuning ($\frac{\Delta_1+\Delta_2}{2}$) and two-photon detuning ($\Delta_1 - \Delta_2$) with and without the RWA. It can be seen from Figures 4a and 4b that under the RWA, the population transfer efficiencies present symmetry with respect to the one-photon and two-photon detunings, and the maximal transfer efficiency is obtained with both the one-photon and two-photon resonances satisfied, which has been shown in references [1–3] as well. In contrast to Figures 4a and 4b, Figure 4c shows that when the pulse areas are about in the low limit of the adiabatic condition, the most efficient population transfer would be realized with the laser frequencies being a little larger than the corresponding transition frequencies and the two-photon resonance almost kept satisfied. However, when the pulse areas are much larger (see Fig. 4d), nearly complete population transfer can take place at several regions of one-photon and two-photon detunings. This is due to the fact that the detunings of the laser fields would lift the quasienergies, and lead to nonadiabatic coupling between the Floquet states [13–16] and subsequent complicated population transfer with respect to the laser detunings. It clearly indicates that the usual one-photon resonances or two-photon resonance is not one necessary condition for maximal population transfer for few-cycle pulses excitation, and tuning the laser frequencies provides an alternative to control of population transfer with much larger Rabi frequencies. Noted that, as seen in Figures 4c and 4d, the population transfer without the RWA can still present robustness to the variations of the one-photon and two-photon detunings in a moderate range.

Under the RWA, the population transfer is irrelevant to the initial optical phases of the laser fields. However, in the case of the few-cycle pulses excitation without the RWA, as the initial optical phase, which is also defined as carrier-envelope phase (CEP) [18,19], determines when the electric-field amplitude peak will appear, the population transfer may show sensitive dependence on the CEP. Figure 5 displays the contour plot of the population transfer efficiency beyond the RWA as a function of the Rabi frequencies and phase difference $\varphi = \varphi_p - \varphi_S$ of the two laser fields with the two fields tuned to resonance with

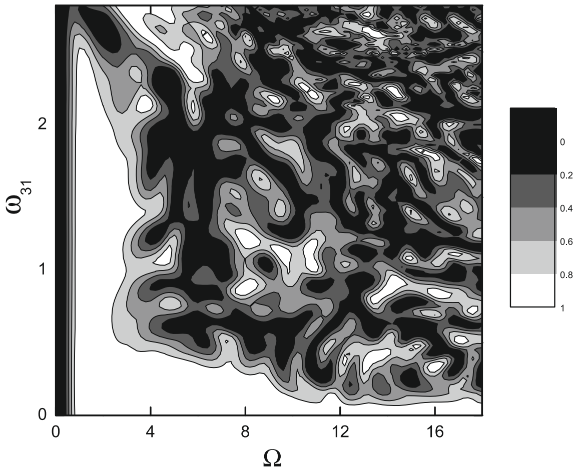

Fig. 6. The contour plot of the population transfer efficiency beyond the RWA as a function of the Rabi frequencies $\Omega = \Omega_{0p} = \Omega_{0S}$ and frequency difference ω_{31} with the two laser fields tuned to resonance with the respective transitions and $\omega_{21} = \pi$, and the other parameters are the same as those in Figure 2.

the respective transitions and the initial phase $\varphi_S = 0$. Unlike the case with the RWA, the population transfer efficiency exhibits periodic distribution with respect to the phase difference of the two fields (with the period equal to π). When the Rabi frequencies of the two fields are about smaller than 2, the transfer efficiency presents almost no dependence on the phase difference, and complete and robust population transfer can be obtained with the Rabi frequencies within the range of about $1.2 \sim 2$. However, when the Rabi frequencies are about larger than 2, the population transfer depends strongly on the Rabi frequencies and phase difference. The variation of the phase difference or Rabi frequencies would lead to low or high transfer efficiency between zero and unity. This means that changing the relative phase of the two laser fields offers an alternative to the control of the population transfer for few-cycle pulses excitation with much larger Rabi frequencies.

Finally, we consider the effect of the frequency difference ω_{31} on the population transfer. Under the RWA, the population transfer efficiency is independence of ω_{31}. However, it is not the case beyond the RWA. Figure 6 shows the contour plot of the population transfer efficiency as a function of the Rabi frequencies and frequency difference ω_{31} with the two laser fields tuned to resonance with the respective transitions, $\omega_{21} = \pi$, and $\varphi_S = \varphi_p = 0$. As seen in Figure 6, when the two lower levels 1 and 3 are nearly degenerate, the population transfer is almost insensitive to the variations of the Rabi frequencies and complete population transfer can be obtained with the Rabi frequencies being about larger than 1.2. In this case, the two pulses excitation beyond the RWA has the similar feature to the piecewise adiabatic passage studied in references [20,21]. When the Rabi frequencies are in the range of about $1.2 \sim 2$, robust and almost perfect population transfer can be achieved with the variations of ω_{31} in the range of about $0 \sim 2$. However, when the Rabi frequencies and ω_{31} are much larger, the intricate behavior of the population transfer efficiency is observed. This is different to the result in reference [14], where each of the two lasers with Rabi frequencies comparable to or larger than the difference of the two transition frequencies interacts with each of the pair of states, and the pulse durations are much larger than optical periods. Detailed calculations show that in order to realize perfect and robust population transfer for the case with large Rabi frequencies and ω_{31}, one can tune the carrier frequencies of the laser fields in a certain range without the need of one-photon or two-photon resonance.

In conclusion, in contrast to the general thought that the most efficient population transfer in the Λ-type three-level system with the STIRAP technique under the RWA would be obtained with the one-photon resonances or two-photon resonance and the initial optical phases of the pump and Stokes laser fields are irrelevant to the population transfer, we demonstrated that the maximal population transfer driven by the few-cycle pump and Stokes laser pulses may be realized without the satisfaction of the one-photon resonances or two-photon resonance and the transfer efficiency depends critically on the Rabi frequencies and initial phases when the peak Rabi frequencies are much larger than the respective transition frequencies. Moreover, complete and robust population transfer can still be obtained with the variations of the Rabi frequencies, pulse durations, and one-photon or two-photon detuning in a moderate range, though a substantial transient population may reside in the excited state. These abnormal results come from the counterrotating terms, which are usually neglected in the rotating wave approximation.

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References

1. K. Bergmann, H. Theuer, B.W. Shore, Rev. Mod. Phys. **70**, 1003 (1998)
2. N.V. Vitanov, T. Halfmann, B.W. Shore, K. Bergmann, Annu. Rev. Phys. Chem. **52**, 763 (2001)
3. N.V. Vitanov, M. Fleischhauer, B.W. Shore, K. Bergmann, Adv. At. Mol. Opt. Phys. **46**, 55 (2001)
4. P. Kral, I. Thanopulos, M. Shapiro, Rev. Mod. Phys. **79**, 53 (2007)
5. X.H. Yang, S.Y. Zhu, Phys. Rev. A **77**, 063822 (2008)
6. X.H. Yang, S.Y. Zhu, Phys. Rev. A **78**, 023818 (2008)
7. M. Kaluza, J.T. Muckerman, Phys. Rev. A **51**, 1694 (1995)
8. L.W. Casperson, Phys. Rev. A **57**, 609 (1998)
9. G.M. Genkin, Phys. Rev. A **58**, 758 (1998)

10. B. Matisov, I. Mazets, L. Windholz, Quantum Semiclass. Opt. **7**, 449 (1995)
11. R.G. Unanyan, S. Guérin, H.R. Jauslin, Phys. Rev. A **62**, 043407 (2000)
12. J. Cheng, J.Y. Zhou, Phys. Rev. A **64**, 065402 (2001)
13. S. Guérin, H.R. Jauslin, Eur. Phys. J. D **2**, 99 (1998)
14. L.P. Yatsenko, B.W. Shore, K. Bergmann, V.I. Romanenko, Eur. Phys. J. D **4**, 47 (1998)
15. R.G. Unanyan, S. Guérin, B.W. Shore, K. Bergmann, Eur. Phys. J. D **8**, 443 (2000)
16. S. Guérin, R.G. Unanyan, L.P. Yatsenko, H.R. Jauslin, Opt. Express **4**, 84 (1999)
17. O.D. Mücke, T. Tritschler, M. Wegener, U. Morgner, F.X. Kärtner, Phys. Rev. Lett. **87**, 057401 (2001)
18. A. Baltuška, T. Udem, M. Uiberacker, M. Hentschel, E. Goulielmakis, C. Gohle, R. Holzwarth, V.S. Yakovlev, A. Scrinzi, T.W. Hänsch, F. Krausz, Nature **421**, 611 (2003)
19. P. Huang, X. Xie, X. Lü, J. Li, X. Yang, Phys. Rev. A **79**, 043806 (2009)
20. E.A. Shapiro, V. Milner, C.M. Jones, M. Shapiro, Phys. Rev. Lett. **99**, 033002 (2007)
21. E.A. Shapiro, A. Pe'er, J. Ye, M. Shapiro, Phys. Rev. Lett. **101**, 023601 (2008)