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Ultrafast control of coherent population transfer with a train of femtosecond pulse pairs

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ABSTRACT

We investigate the ultrafast control of coherent population transfer in a Λ-type three-level system with a train of pump–Stokes femtosecond pulse pairs, where the pulse sequences can be produced either by optical delay line or by pulse shaping with sinusoidal phase modulation. It is shown that when the pump and Stokes pulses in each pair are applied in the counterintuitive order, similar to that in the stimulated Raman adiabatic passage technique, due to temporal quantum interference (besides optical interference in the case of overlapped subpulses), ultrafast control of coherent population transfer can be achieved by scanning the inter-pair time delay or by changing the sinusoidal phase modulation parameters. This method has potential applications in ultrafast control of chemical reactions and quantum information processing.

Much attention has been paid to quantum coherent control of an atomic or a molecular system from an initial state to a particular target state in recent years. The underlying physical mechanism of coherent control is quantum coherence and interference. As is well-known, there exist two kinds of quantum interferences. One refers to the interference between two or more different transition pathways connecting the same initial or final state in the frequency domain (usually referred to as the "Brumer–Shapiro" scenario) [1–3], and the other refers to the interference between two or more time-separated transition pathways connecting the same initial and final states in the time domain [4–8]. Three main strategies, that is, temporal coherent control (TCC), optimal control (OC), and adiabatic passage (AP), have been proposed to realize quantum coherent control [9]. TCC uses temporal quantum interference to achieve selectivity of population transfer [5–8]; OC employs pulse-shaping technique to optimize the laser pulse for controlling population transfer [10–13], and AP can realize selectivity and completeness of population transfer between two quantum states, such as stimulated Raman adiabatic passage (STIRAP) and Stark-chirped rapid adiabatic passage [14–17]. Apart from exerting one of the above-mentioned methods, the combination of two of them has also been exploited recently to realize quantum control. Shapiro et al. [18,19] developed a method of executing complete population transfer between quantum states by merging AP and pulse-shaping techniques. Baumert [20] combined the strategies of TCC and pulse shaping to realize the control of sodium atomic two-photon transitions, and Kral and Shapiro [9] proposed a way of combining AP and coherent control, termed as coherently controlled adiabatic passage (CCAP), to achieve both selectivity and completeness of population transfer. By using a shaped fs frequency comb formed by a train of fs mode-locked pulses, Ye et al. [21,22] presented an efficient scheme for precise control of molecular dynamics and atomic two-photon transition. Recently, we have studied the ultrafast coherent population transfer in a Λ-type three-level system driven by a train of weak pump–Stokes femtosecond pulse pairs with two pulse sequences having the same optical phases [23]. In this paper, we investigate the ultrafast control of coherent population transfer in a Λ-type three-level system with a train of pump–Stokes fs pulse pairs by combining TCC and pulse-shaping techniques, where the pulse sequences can be either the phase-locked interferometrically-generated pulses through optical delay line or the sinusoidally phase-modulated pulses through pulse shaping. By scanning the inter-pair time delay or by changing the sinusoidal spectral phase modulation

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parameters, ultrafast control of coherent population transfer can be realized with the pump and Stokes pulses in each pair applied in the counterintuitive order, similar to that in the STIRAP technique.

The considered Λ-type three-level system, as shown in Fig. 1, interacts with a train of pump–Stokes fs laser pulse pairs, where level 1 and upper level 2 is coupled by the pump pulse train, and levels 2 and 3 is coupled by the Stokes pulse train. Firstly, we consider the pump and Stokes pulse sequences are the phase-locked interferometrically-generated pulses, which can be experimentally produced by the same laser pulse with an optical delay line or an optical cavity, as discussed by Salour [5] and Vitanov and Knight [24]. For simplicity, we assume that both the pump and Stokes pulse trains are formed by N equally-spaced pulses with the same amplitude envelopes and peak values. The electric field $E_S(t)$ of the Stokes pulse train can be written as $E_S(t) = \sum_{n=0}^{N} E_0 f_S(t - nT)\cos[\omega_S(t - nT)]$, where ω_S is the central frequency of the Stokes field, $f_S(t - nT)$ is the electric-field amplitude envelope of the nth Stokes pulse with the peak value of E_0, and T is the repetition period of the Stokes pulse train. We assume the time delay between the first pump–Stokes pair is τ, the first pair has the same initial optical phase, and the time origin is chosen at the peak of the first Stokes pulse, so the electric field $E_p(t)$ of the pump pulse train can be written as $E_p(t) = \sum_{n=0}^{N} E_0 f_p(t - \tau - nT')\cos[\omega_p(t - nT')]$ with the repetition period of the pump pulse train being T'. The Rabi frequencies of the nth Stokes and pump pulses are set to be Gaussian with the amplitude envelopes of the form $\Omega_{nS}(t) = \Omega_{0S}\exp[-(t - nT)^2/T_p^2]$ and $\Omega_{np}(t) = \Omega_{0p}\exp[-(t - \tau - nT')^2/T_p^2]$, respectively, where T_p is the pulse duration, $\Omega_{0p}(S) = \mu_{1(2)} \cdot E_0/\hbar$ is the peak value of the Rabi frequency of the pump (Stokes) pulse, with μ_1 (μ_2) being the dipole moment for the transition 1–2 (2–3). For simplicity, we assume that Ω_{0p} and Ω_{0S} are equal to each other, and both laser fields are tuned to resonance with the respective transitions. We neglect the population decay from upper level 2, as the interaction time (about tens to hundreds of fs) is far smaller than the population decay time (about tens of ns). In the rotating-wave approximation, the time-dependent Hamiltonian of the atom–field system can be written as

$$H = \hbar\begin{pmatrix} 0 & \Omega_p^*(t) & 0 \\ \Omega_p(t) & 0 & \Omega_S(t) \\ 0 & \Omega_S^*(t) & 0 \end{pmatrix} \quad (1)$$

In the above equation, $\Omega_S(t)$ and $\Omega_p(t)$ are the Rabi frequencies of the Stokes and pump pulse trains, respectively. The time evolution of the system can be readily treated by resolving the time-dependent Schrodinger equation with the fourth-order Runge–Kutta integrator. In what follows, the population is initially in level 1, and the relevant parameters are scaled with fs (or fs^{-1}).

Fig. 2 displays the time evolution of the populations in the three states with both the pump and Stokes laser fields tuned to resonance with the respective transitions under different inter-pair time delay T of the Stokes pulse train with $\Omega_{0p} = \Omega_{0S} = 0.2\ fs^{-1}$, $T_p = 30$ fs, $\tau = 40$ fs, and $\omega_S = \omega_p = \pi$ rad/fs or $\omega_S = 0.8\omega_p = 0.8\pi$ rad/fs for two pairs of phase-locked pump–Stokes pulses excitation

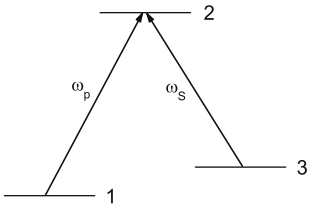

Fig. 1. The Λ-type three-level system driven by a train of pump–Stokes laser pulse pairs with the central frequencies ω_p and ω_S, respectively.

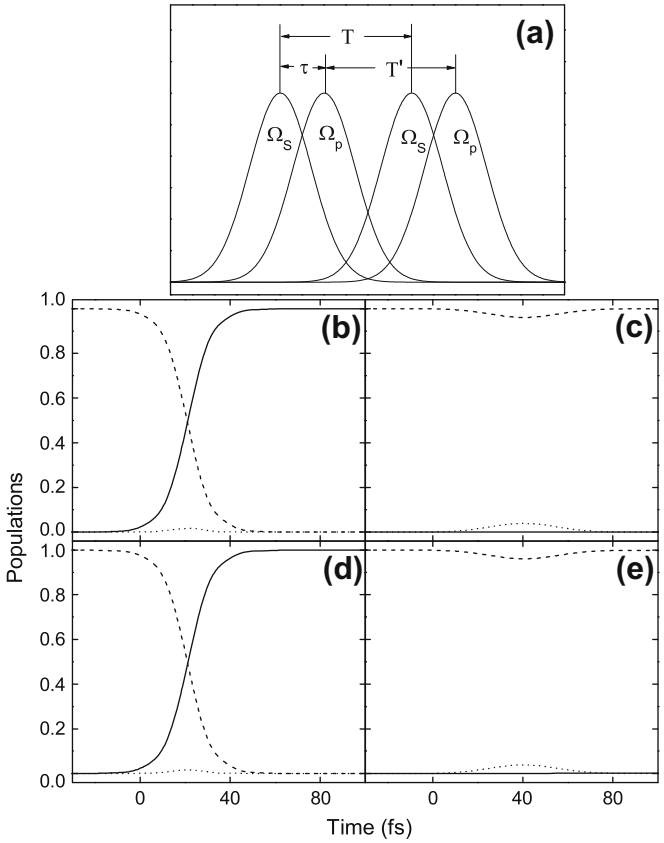

Fig. 2. (a) The pump and Stokes pulse sequences formed by two pulse pairs with the first intrapair time delay τ and inter-pair time delays T and T'. (b–e) The time evolution of the populations in the states 1 (dashed line), 2 (dotted line), and 3 (solid line) with both the pump and Stokes laser fields tuned to resonance with the respective transitions with $\Omega_{0p} = \Omega_{0S} = 0.2\ fs^{-1}$, $T_p = 30$ fs, $\tau = 40$ fs, $\omega_S T = \omega_p T'$, $\omega_S = \omega_p = \pi$ rad/fs (b and c) and $\omega_S = 0.8\omega_p = 0.8\pi$ rad/fs (d and e) for $T = 0$ fs (b and d), and $T = 2$ fs (c and e).

(shown in Fig. 2a). In order to have a fixed relative optical phase between each pump–Stokes pair, we set $\omega_S T = \omega_p T'$. In this case, the Rabi frequencies $\Omega_p(t)$ and $\Omega_S(t)$ in Eq. (1) can be written as $\Omega_p(t) = \sum_{n=0}^{N}\Omega_{np}(t)\exp(jn\omega_p T')$ and $\Omega_S(t) = \sum_{n=0}^{N}\Omega_{nS}(t)\exp(jn\omega_S T)$, respectively. Note that for the degenerate case $\omega_S = \omega_p = \pi$ rad/fs, that each laser field interacts with only one pair of states can be realized with the photons of different polarization from a same laser beam [25]. As seen from Fig. 2b and d, when the inter-pair time delay T is equal to zero, which means the two pulses in each train are completely overlapped, nearly complete population transfer can be obtained via the STIRAP process in both cases $\omega_S = \omega_p = \pi$ rad/fs and $\omega_S = 0.8\omega_p = 0.8\pi$ rad/fs. However, for the case of the inter-pair time delay $T = 2$ fs (see Fig. 2c and e), due to temporal destructive quantum interference besides optical interference, almost no population transfer from state 1 to state 3 can take place, and only a small transient population would reside in the excited state 2 during the evolution process.

In order to see how the inter-pair time delay T can control population transfer, we show in Fig. 3 the final populations in the three states 1, 2, and 3 as a function of T for two pairs of phase-locked pump–Stokes pulses excitation for the two cases $\omega_S = \omega_p = \pi$ rad/fs and $\omega_S = 0.8\omega_p = 0.8\pi$ rad/fs. Obviously, as seen in Fig. 3a, ultrafast control of population transfer can be obtained by varying the inter-pair time delay T. When $T = 0$ fs, perfect population transfer can be obtained. With the increase of T, the transfer efficiency exhibits fast oscillations with the period of 2 fs