Analytic properties and effective two-level problems in stimulated Raman adiabatic passage

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We demonstrate that various properties of population transfer by delayed pulses in three-level systems on two-photon resonance can be deduced analytically and for general pulse shapes. We use the fact that the three-level system reduces to effective two-level problems at large intermediate-level detuning Δ, on resonance ($\Delta=0$) and for completely overlapping pulses. Special attention is paid to the effect of the pulse order on the population transfer efficiency. We show that on resonance the transfer efficiency depends substantially on the pulse order, while at large Δ it does not. We also find that under some natural restrictions on the symmetry of the problem, the population of the initial level does not depend on the pulse order at any Δ. Furthermore, we demonstrate that the population transfer in the three-level system can be viewed as a level-crossing problem in an equivalent two-level system not only at large Δ (which is known) but also on resonance, $\Delta=0$. The effective on-resonance two-level problem is interesting by itself as it shows that a level crossing and adiabatic evolution do not necessarily lead to complete population inversion. As examples throughout the paper, we present several analytically solvable models. [S1050-2947(97)09301-3]

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I. INTRODUCTION

In recent years, the stimulated Raman adiabatic passage process (STIRAP) has been intensively studied experimentally, analytically, and numerically [1–13]. On the one hand, this process provides the possibility for efficient population transfer in three-level and even multilevel systems by using relatively simple experimental setups. On the other, it gives an interesting example of counterintuitive physics. In a three-level Λ system (Fig. 1), STIRAP requires that the Stokes pulse Ω_s, driving the transition between the initially unpopulated levels 2 and 3, and *precedes* the pump pulse Ω_p, which drives the transition between the initially populated level 1 and the intermediate level 2, though they overlap partly. This is the so-called *counterintuitive* pulse sequence, in contrast to the *intuitive* one in which the pump pulse Ω_p precedes the Stokes pulse Ω_s. STIRAP exploits the existence of an eigenstate of the Hamiltonian, which involves states 1 and 3 only. Such an eigenstate appears when the Λ system is on *two-photon resonance*, a condition which we will assume throughout the paper. The intermediate level 2 can be off resonance by a detuning Δ.

The properties of the population transfer in three-level systems have mainly been deduced from numerical simulations [2–5]. Analytically, the process has been treated either in the perfect adiabatic limit [6–9] or for specific pulse shapes on resonance [5,10,11]. In this paper, we demonstrate that various properties can be derived analytically and for general pulse shapes, with a particular emphasis on the effect of the pulse order on the population transfer efficiency. Most of these properties have not been discussed in the literature so far, to our knowledge.

The paper is organized as follows. In Sec. II, we present the basic equations and definitions. In Sec. III, we analyze the population transfer in the adiabatic limit. Beyond the adiabatic limit, the analysis is more difficult but, fortunately, it is simplified by the fact that the three-level system reduces to effective two-level problems on resonance ($\Delta=0$), at large intermediate-level detuning Δ and for completely overlapping pulses; these two-level problems are presented in Sec. IV. In Sec. V, we demonstrate that STIRAP can be viewed as a level-crossing problem in an equivalent two-level system not only at large Δ (which is known [7,13]) but also on resonance. We also consider the effective on-resonance level-crossing problem by itself as it appears to be quite unusual. Finally, in Sec. VI, we summarize the conclusions.

II. THREE-LEVEL SYSTEM

Consider the three-level Λ system shown schematically in Fig. 1. Levels 1 and 2 are coupled by the pump laser pulse $\Omega_p(t)$, while levels 2 and 3 are coupled by the Stokes laser pulse $\Omega_s(t)$. The transition between levels 1 and 3 is electric dipole forbidden. Two-photon resonance between levels 1 and 3 is maintained, while level 2 can be off resonance by a certain detuning Δ. The wave functions of these levels (the bare states) will be denoted by $|1\rangle$, $|2\rangle$, and $|3\rangle$. The pulse durations are supposed to be short compared with the relaxation times of the system. Then the time evolution of the probability amplitudes $\mathbf{c}(t)=[c_1(t),c_2(t),c_3(t)]^T$ of the three states is governed by the Schrödinger equation (in units $\hbar=1$)

$$i\frac{d}{dt}\mathbf{c}(t)=\mathbf{H}(t)\mathbf{c}(t), \tag{1}$$

where the Hamiltonian of the system in the rotating-wave approximation is given by

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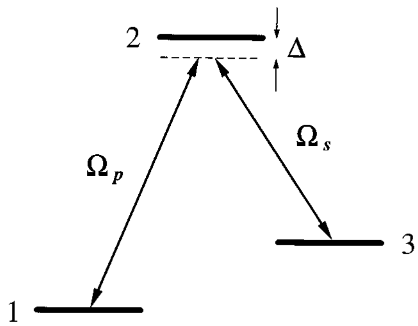

FIG. 1. The three-level Λ system. Levels 1 and 2 are coupled by the pump laser pulse $\Omega_p(t)$, while levels 2 and 3 are coupled by the Stokes laser pulse $\Omega_s(t)$. The transition between levels 1 and 3 is electric dipole forbidden. Levels 1 and 3 are on two-photon resonance, while level 2 may be off resonance by a certain detuning Δ. Only level 1 is populated initially. In STIRAP the Stokes pulse $\Omega_s(t)$ precedes the pump pulse $\Omega_p(t)$ (counterintuitive pulse order).

$$\mathbf{H}(t)=\begin{bmatrix} 0 & \Omega_p(t) & 0 \\ \Omega_p(t) & \Delta(t) & \Omega_s(t) \\ 0 & \Omega_s(t) & 0 \end{bmatrix}. \tag{2}$$

Without loss of generality the detuning Δ(t) and the functions $\Omega_p(t)$ and $\Omega_s(t)$, representing the Rabi frequencies of the two pulses, will be assumed positive. Furthermore, $\Omega_p(t)$ and $\Omega_s(t)$ are supposed to be pulse-shaped functions, that is, functions which vanish at infinity and whose pulse areas $\int_{-\infty}^{\infty}\Omega_{p,s}(t)dt$ are finite. Generally, we will not impose any specific restrictions on $\Omega_p(t)$ and $\Omega_s(t)$ regarding pulse shapes, symmetries, and so on. In some cases, which will be explicitly stated, considerable simplifications occur if we choose

$$\Omega_p(t)=\frac{\alpha}{T}f\left(\frac{t-\tau}{T}\right), \quad \Omega_s(t)=\frac{\alpha}{T}f\left(\frac{t+\tau}{T}\right), \tag{3}$$

where $f(x)=f(-x)$ is a symmetric pulse-shaped function, i.e., if the pulses have the same strength, the same shape described by $f(x)$, and the same characteristic width T. Here 2τ is the delay between the pulses, and $\tau<0$ means intuitive pulse order while $\tau>0$ means counterintuitive pulse order.

We suppose that at time $t\rightarrow-\infty$ the three-level system is, in state $|1\rangle$,

$$c_1(-\infty)=1, \quad c_2(-\infty)=0, \quad c_3(-\infty)=0, \tag{4}$$

and we are interested in the populations at time $t\rightarrow+\infty$, $P_n=|c_n(+\infty)|^2$ ($n=1$, 2, and 3).

The population transfer mechanism is most easily revealed in the adiabatic representation, i.e., in the basis of the instantaneous eigenstates $|+\rangle$, $|0\rangle$, and $|-\rangle$ of $\mathbf{H}(t)$, called adiabatic states. They are connected to the bare (diabatic) states $|1\rangle$, $|2\rangle$, and $|3\rangle$ by the relations [5]

$$\begin{aligned} |+\rangle &= \sin\varphi\,\sin\vartheta|1\rangle+\cos\varphi|2\rangle+\sin\varphi\,\cos\vartheta|3\rangle, \\ |0\rangle &= \cos\vartheta|1\rangle-\sin\vartheta|3\rangle, \\ |-\rangle &= \cos\varphi\,\sin\vartheta|1\rangle-\sin\varphi|2\rangle+\cos\varphi\,\cos\vartheta|3\rangle. \end{aligned} \tag{5}$$

These adiabatic states correspond to the eigenvalues of $\mathbf{H}(t)$,

$$\lambda_+(t)=\tfrac{1}{2}[\Delta(t)+\sqrt{\Delta^2(t)+4\Omega_0^2(t)}]=\Omega_0(t)\cot\varphi(t),$$

$$\lambda_0(t)=0,$$

$$\lambda_-(t)=\tfrac{1}{2}[\Delta(t)-\sqrt{\Delta^2(t)+4\Omega_0^2(t)}]=-\Omega_0(t)\tan\varphi(t),$$

with the time-dependent Euler's angles ϑ and φ defined as

$$\tan\vartheta(t)=\frac{\Omega_p(t)}{\Omega_s(t)}, \tag{6}$$

$$\tan 2\varphi(t)=\frac{2\Omega_0(t)}{\Delta(t)}, \tag{7}$$

and

$$\Omega_0(t)=\sqrt{\Omega_p^2(t)+\Omega_s^2(t)}. \tag{8}$$

Likewise, the probability amplitudes of the adiabatic states $\mathbf{a}(t)=[a_+(t),a_0(t),a_-(t)]^T$ are connected to the diabatic (bare) amplitudes $\mathbf{c}(t)$ by [5]

$$\mathbf{c}(t)=\mathbf{W}(t)\mathbf{a}(t), \tag{9}$$

where the orthogonal rotation matrix $\mathbf{W}(t)$ is given by

$$\mathbf{W}(t)=\begin{bmatrix} \sin\varphi\,\sin\vartheta & \cos\vartheta & \cos\varphi\,\sin\vartheta \\ \cos\varphi & 0 & -\sin\varphi \\ \sin\varphi\,\cos\vartheta & -\sin\vartheta & \cos\varphi\,\cos\vartheta \end{bmatrix}. \tag{10}$$

The Schrödinger equation in the adiabatic representation is obtained from Eqs. (1), (9), and (10) and is given by

$$i\frac{d}{dt}\mathbf{a}(t)=\mathbf{H}_a(t)\mathbf{a}(t), \tag{11}$$

with

$$\mathbf{H}_a(t)=\begin{bmatrix} \Omega_0\cot\varphi & i\dot{\vartheta}\,\sin\varphi & i\dot{\varphi} \\ -i\dot{\vartheta}\,\sin\varphi & 0 & -i\dot{\vartheta}\,\cos\varphi \\ -i\dot{\varphi} & i\dot{\vartheta}\,\cos\varphi & -\Omega_0\tan\varphi \end{bmatrix}, \tag{12}$$

where an overdot means a time derivative.

In Sec. III we discuss the population transfer mechanisms for the two pulse orders in the adiabatic limit, and the conditions for adiabatic evolution. Generally, adiabatic evolution is achieved for large pulse strengths and/or large pulse widths; hence the product of the pulse strength and the pulse width, which is proportional to the pulse area and will be denoted by α, usually serves as the adiabaticity parameter (the larger α is, the stronger the adiabaticity).

III. ADIABATIC LIMIT

A. Counterintuitive pulse order

The population transfer by pulses in *counterintuitive* order [in which the Stokes pulse $\Omega_s(t)$ precedes the pump pulse $\Omega_p(t)$], referred to as STIRAP, exploits the existence of the adiabatic state $|0\rangle$ which is a time-dependent linear combination of states $|1\rangle$ and $|3\rangle$ only. The counterintuitive order means that

$$\lim_{t\to-\infty}\frac{\Omega_p(t)}{\Omega_s(t)}=0,\quad \lim_{t\to+\infty}\frac{\Omega_p(t)}{\Omega_s(t)}=\infty, \tag{13}$$

or, in other words,

$$\vartheta^{\rm ci}(-\infty)=0,\quad \vartheta^{\rm ci}(+\infty)=\frac{\pi}{2}. \tag{14}$$

Hence the adiabatic state $|0\rangle$ is equal to state $|1\rangle$ before excitation and to state $|3\rangle$ after it, so that among the adiabatic states only $|0\rangle$ is populated initially. If the excitation is adiabatic, the system will remain in this adiabatic state all the time and, ultimately, the population will be completely transferred to state $|3\rangle$,

$$P_1^{\rm ci}(+\infty)\approx 0,\quad P_2^{\rm ci}(+\infty)\approx 0,\quad P_3^{\rm ci}(+\infty)\approx 1 \tag{15}$$

Here and in what follows a superscript ''ci'' (''i'') indicates a quantity related to the counterintuitive (intuitive) pulse order. A remarkable property of the three-level system on two-photon resonance is that the adiabatic state $|0\rangle$ does not involve the intermediate state $|2\rangle$. This implies that if the evolution is nearly adiabatic, the population of level $|2\rangle$ will remain negligible throughout the excitation and, thus, the specific properties of state $|2\rangle$, including possible decay to other states, should not substantially influence the efficiency of STIRAP. This is an important advantage of STIRAP compared with the other population transfer mechanisms. Finally, we should note that in the adiabatic limit, the value of the intermediate-level detuning Δ does not affect the transfer efficiency as the adiabatic state $|0\rangle$ does not depend on it. Beyond the adiabatic limit, however, the detuning Δ does affect the transfer efficiency. This interesting issue, which we will not consider in this work, has been studied in our recent paper [12].

B. Intuitive pulse order

Consider now the *intuitive* pulse order in which the pump pulse $\Omega_p(t)$ precedes the Stokes pulse $\Omega_s(t)$. In other words,

$$\lim_{t\to-\infty}\frac{\Omega_p(t)}{\Omega_s(t)}=\infty,\quad \lim_{t\to+\infty}\frac{\Omega_p(t)}{\Omega_s(t)}=0, \tag{16}$$

which means that

$$\vartheta^{\rm i}(-\infty)=\frac{\pi}{2},\quad \vartheta^{\rm i}(+\infty)=0. \tag{17}$$

Unlike the counterintuitive pulse order, there is a substantial difference between the cases of $\Delta=0$ and $\Delta\neq 0$.

(i) For $\Delta=0$, we have $\varphi\equiv\pi/4$, which implies that, initially, the adiabatic states $|+\rangle$ and $|-\rangle$ are both populated while the state $|0\rangle$ is not populated, as $a_+(-\infty)=a_-(-\infty)=1/\sqrt{2}$, $a_0(-\infty)=0$ [see Eqs. (4) and (9)]. In the adiabatic limit, the final adiabatic amplitudes are $a_+(+\infty)=a_-^*(+\infty)=e^{i\zeta}/\sqrt{2}$, $a_0(+\infty)=0$, where ζ is the adiabatic phase,

$$\zeta=\int_{-\infty}^{\infty}\Omega_0(t)dt. \tag{18}$$

From Eq. (9) we find that the populations of the bare (diabatic) states are

$$P_1^{\rm i}(+\infty)\approx 0,\quad P_2^{\rm i}(+\infty)\approx\sin^2\zeta,\quad P_3^{\rm i}(+\infty)\approx\cos^2\zeta. \tag{19}$$

Thus, the final-state population $P_3^{\rm i}(+\infty)$ is not equal to unity, as for the counterintuitive pulse order, but oscillates with the adiabaticity parameter since ζ is proportional to it [if Eq. (3) is assumed then α plays the role of the adiabaticity parameter].

(ii) For $\Delta\neq 0$, we have $\varphi(-\infty)=\varphi(+\infty)=0$, which implies that only state $|-\rangle$ among the adiabatic states is populated initially as $a_+(-\infty)=a_0(-\infty)=0$, $a_-(-\infty)=1$ [see Eqs. (4) and (9)]. Furthermore, state $|-\rangle$ coincides with state $|3\rangle$ at $t\to+\infty$; thus, if the excitation is adiabatic, then the system will remain in state $|-\rangle$ and the population will eventually be completely transferred to state $|3\rangle$. Therefore, *in the adiabatic limit both the intuitive and counterintuitive pulse orders produce complete population transfer for nonzero intermediate-level detuning,* $\Delta\neq 0$. There is, however, a difference in the way the population is transferred from state $|1\rangle$ to state $|3\rangle$. For the counterintuitive pulse order, the population is transferred through the adiabatic state $|0\rangle$ which does not involve the intermediate state $|2\rangle$ and, thus, no population visits state $|2\rangle$ at any time. For the intuitive pulse order, the population is transferred through the adiabatic state $|-\rangle$ which involves state $|2\rangle$ [see Eqs. (5)]. Thus, the intermediate state $|2\rangle$ *is* populated during the transfer and in the adiabatic limit its population is

$$P_2^{\rm i}(t)\approx\sin^2\varphi(t)=\frac{1}{2}\left[1-\frac{\Delta(t)}{\sqrt{4\Omega_0^2(t)+\Delta^2(t)}}\right].$$

The maximum value of $P_2^{\rm i}(t)$ is in the interval $(0,\frac{1}{2})$, and it is determined by the particular case considered. This means that, in the case of strong decay from the intermediate state, the counterintuitive pulse order is again advantageous as compared with the intuitive pulse order. Related results have earlier been obtained for a four-level system in [9].

C. An example: Gaussian pulses

To illustrate the above conclusions, we integrated Eqs. (1) numerically in the case of Gaussian pulses of the same shapes and strengths but separated by a pulse delay of 2τ,

$$\Omega_p(t)=\frac{\alpha}{T}\exp\left[-\left(\frac{t-\tau}{T}\right)^2\right],\quad \Omega_s(t)=\frac{\alpha}{T}\exp\left[-\left(\frac{t+\tau}{T}\right)^2\right],$$

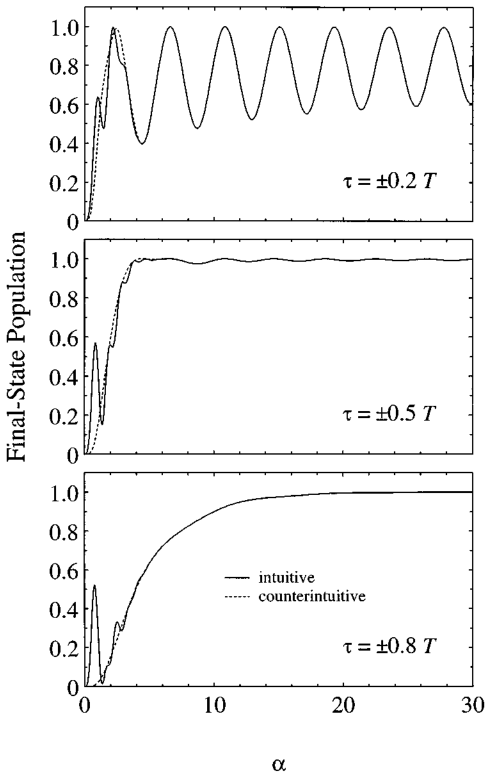

FIG. 2. The final-state population for Gaussian pulses (20) plotted as a function of the dimensionless parameter α for $\tau=\pm 0.2T$, $\pm 0.5T$, and $\pm 0.8T$ ($\tau>0$ means the counterintuitive pulse order, while $\tau<0$ means the intuitive order). The detuning δ is always set equal to α.

$$\Delta(t)=\frac{\delta}{T}, \tag{20}$$

where α, δ, and T are positive parameters, while τ can be positive or negative. A positive τ means a counterintuitive pulse order, while a negative τ means an intuitive order. The parameters α and δ are dimensionless, while T and τ have the dimension of time. In Fig. 2, the final-state population is plotted as a function of α for $\tau=\pm 0.2T$, $\pm 0.5T$, and $\pm 0.8T$. The detuning δ is set equal to α. The figure demonstrates how the adiabatic limit is approached for both counterintuitive and intuitive pulse orders when the adiabaticity increases. For small τ ($\tau=\pm 0.2T$), the adiabatic limit (unity transfer efficiency) is approached slowly and in an oscillatory manner because of too much overlap between the pulses which leads to large-amplitude Rabi oscillations (see Sec. IV C). For large τ ($\tau=\pm 0.8T$), there are almost no oscillations, but the adiabatic limit is approached slowly because of too small overlap between the pulses which requires large pulse strengths to achieve adiabaticity. From the point of view of the transfer efficiency, the region of moderate pulse separation ($\tau=\pm 0.5T$) is the optimal one. We also see in the figure that in all cases, a difference between the intuitive and counterintuitive orders exists only at small α (and this means at small δ too), i.e., away from the adiabatic regime and near resonance. For large α (and this means for large δ), both pulse orders produce almost the same transfer efficiency; this feature is discussed in more detail in Sec. IV B.

As we mentioned, in Fig. 2, δ ($\equiv \Delta T$) is set equal to α ($\equiv \Omega_{p,\max}T=\Omega_{s,\max}T$), i.e., δ and α increase simultaneously. This can be achieved either by increasing simultaneously the detuning Δ and the pulse amplitudes $\Omega_{p,\max}$ and $\Omega_{s,\max}$ for fixed pulse widths T or, alternatively, by increasing the pulse widths T for fixed detuning and pulse amplitudes. The reason is that for the intuitive pulse order, adiabaticity cannot be achieved by simply increasing α for a fixed δ. This is not a problem for the counterintuitive pulse order because then the population transfer is realized through the adiabatic state $|0\rangle$, which is coupled to the other adiabatic states by the matrix elements containing $\dot{\vartheta}$ [see Eq. (12)]. The diagonal elements $\Omega_0\cot\varphi$ and $-\Omega_0\tan\varphi$ can always be made much larger than the matrix elements with $\dot{\vartheta}$ for sufficiently large α. For the intuitive pulse order, however, the population transfer is realized through the adiabatic state $|-\rangle$, which is coupled to the adiabatic state $|+\rangle$ by the off-diagonal matrix elements with $\dot{\varphi}(t)$. These latter elements possess two peaks situated on the (rapidly vanishing) wings of $\Omega_0(t)$, one for $t<0$ and another for $t>0$. As α increases, these peaks move away from the pulses and, hence, if in these regions $\dot{\varphi}$ is of the order of or larger than Ω_0, nonadiabatic transitions take place which deteriorate the transfer efficiency. Thus we cannot eliminate this nonadiabatic coupling by increasing α alone. If δ is increased simultaneously with α, so that the ratio α/δ is kept constant, then $\dot{\varphi}$ does not change; thus, for sufficiently large α (and δ), we can achieve $\Omega_0 \gg |\dot{\varphi}|$ at all times. We hence conclude that *the adiabatic regime is easier to approach for counterintuitive pulses than it is for intuitive pulses*.

In Fig. 3, we show the time evolution of the populations for counterintuitive (upper figure) and intuitive (lower figure) pulse orders with $\alpha=\delta=10$ and $\tau=\pm 0.5T$. These values of the parameters ensure nearly adiabatic evolution. The figure demonstrates that although an almost complete population transfer to the final state is realized in both cases, for the counterintuitive order the population of the intermediate level 2 remains very small during the excitation, while for the intuitive order it reaches appreciable values.

IV. EQUIVALENT TWO-LEVEL PROBLEMS AND PULSE ORDER EFFECTS *BEYOND* THE ADIABATIC LIMIT

Neither Eqs. (1) nor (11) are easy to analyze analytically *beyond the adiabatic limit*. Fortunately, in three important limits—on resonance ($\Delta=0$), at large detuning ($\Delta \gg \Omega_p, \Omega_s$), and for completely overlapping pulses—the three-level problem is reduced to effective two-level problems which greatly facilitate the analysis.

A. On resonance

1. Effective two-level problem

In the case of intermediate-level resonance, $\Delta=0$, the three-level problem is reduced to an equivalent two-level one with a Rabi frequency $\frac{1}{2}\Omega_p(t)$ and a detuning $\frac{1}{2}\Omega_s(t)$. This is possible because, for $\Delta=0$, Eqs. (1) have the same form as the optical Bloch equations for a two-level system. The

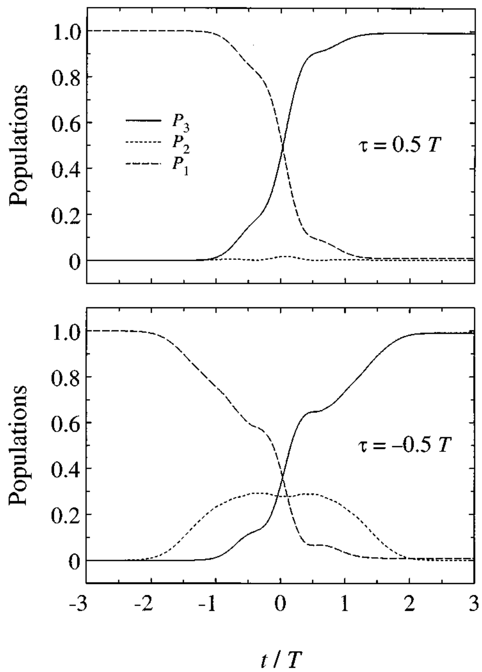

FIG. 3. The time evolution of the populations for counterintuitive (upper figure) and intuitive (lower figure) pulse orders for Gaussian pulses (20) with $\alpha=\delta=10$ and $\tau=0.5T$ and $\tau=-0.5T$, respectively.

Schrödinger equation for the probability amplitudes $\mathbf{b}(t)=[b_1(t),b_2(t)]^T$ of the effective two-level system reads [5]

$$i\frac{d}{dt}\mathbf{b}(t)=\frac{1}{2}\begin{bmatrix}-\Omega_s(t) & \Omega_p(t)\\ \Omega_p(t) & \Omega_s(t)\end{bmatrix}\mathbf{b}(t). \tag{21}$$

By using the transformation

$$\mathbf{b}(t)=\mathbf{R}[\tfrac{1}{2}\,\vartheta(t)]\mathbf{d}(t) \tag{22}$$

to the adiabatic amplitudes $\mathbf{d}(t)=[d_1(t),d_2(t)]^T$, where $\vartheta(t)$ is defined by Eq. (6) and

$$\mathbf{R}(\phi)=\begin{bmatrix}\cos\phi & \sin\phi\\ -\sin\phi & \cos\phi\end{bmatrix}, \tag{23}$$

the two-state equations (21) are transformed into the adiabatic equations

$$i\frac{d}{dt}\mathbf{d}(t)=\frac{1}{2}\begin{bmatrix}-\Omega_0(t) & -i\dot{\vartheta}(t)\\ i\dot{\vartheta}(t) & \Omega_0(t)\end{bmatrix}\mathbf{d}(t). \tag{24}$$

Apparently, the adiabaticity condition for Eqs. (24), $|\dot{\vartheta}|\ll\Omega_0$, is identical to that for Eqs. (11) with $\Delta=0$. The probability amplitudes $\mathbf{c}(t)$ of the three-level system are related to the bare two-level amplitudes $\mathbf{b}(t)$ by

$$c_1(t)=1-2|b_2(t)|^2,\quad c_2(t)=-2i\ \mathrm{Im}[b_1(t)b_2^*(t)],$$
$$c_3(t)=2\ \mathrm{Re}[b_1(t)b_2^*(t)], \tag{25}$$

and to the adiabatic two-level amplitudes $\mathbf{d}(t)$ by

$$c_1(t)=(|d_1(t)|^2-|d_2(t)|^2)\cos\vartheta(t)+2\mathrm{Re}[d_1(t)d_2^*(t)]\sin\vartheta(t),$$
$$c_2(t)=-2i\ \mathrm{Im}[d_1(t)d_2^*(t)],$$
$$c_3(t)=2\ \mathrm{Re}[d_1(t)d_2^*(t)]\cos\vartheta(t)-(|d_1(4)|^2-|d_2(t)|^2)\sin\vartheta(t). \tag{26}$$

These relations hold for any pulse order.

2. Intuitive and counterintuitive pulse orders

The relation between the three-level on-resonance problem and the effective two-level problem enables us to show explicitly that, as the adiabaticity increases, the transfer efficiency for the counterintuitive pulse sequence approaches unity, while it oscillates for the intuitive pulse sequence. Let $\Omega_1(t)$ and $\Omega_2(t)$ be two delayed but partly overlapping pulses with $\Omega_1(t)$ preceding $\Omega_2(t)$,

$$\lim_{t\to-\infty}\frac{\Omega_1(t)}{\Omega_2(t)}=\infty,\quad \lim_{t\to+\infty}\frac{\Omega_1(t)}{\Omega_2(t)}=0. \tag{27}$$

We will consider the special case when the pulse order is reversed by simply interchanging the pump and Stokes pulses. In other words, in the counterintuitive pulse sequence we take $\Omega_p(t)=\Omega_2(t)$ and $\Omega_s(t)=\Omega_1(t)$, while in the intuitive pulse sequence we set $\Omega_p(t)=\Omega_1(t)$ and $\Omega_s(t)=\Omega_2(t)$. Then the Hamiltonians in Eqs. (21) [as well as in Eqs. (1)] corresponding to the two pulse orders are related to each other by a simple time-independent unitary transformation; so are the respective evolution matrices. This implies that the solutions for the two pulse orders can be expressed in terms of *the same* interaction parameters. The same conclusion can be drawn if one starts from the adiabatic equations (24). Interchanging Ω_1 and Ω_2 does not change anything in these equations but the sign of $\dot{\vartheta}$. Consequently, the evolution matrices are the same except for the signs of the off-diagonal elements. In this case, the substantial difference between the two pulse orders comes from the different initial conditions $\mathbf{d}(-\infty)$ [because $\vartheta(-\infty)$ is different] which are determined from Eqs. (26) and are imposed in order to satisfy Eqs. (4). Therefore, the solutions $\mathbf{d}^{\mathrm{i}}(+\infty)$ and $\mathbf{d}^{\mathrm{ci}}(+\infty)$ for the two pulse orders can be obtained from essentially the same evolution matrix [though applied on different initial vectors $\mathbf{d}(-\infty)$], i.e., in terms of the same interaction parameters.

It is most convenient to express the solutions for the two pulse orders in terms of the parameters of the evolution matrix $\mathbf{U}_d(+\infty,-\infty)$ for the adiabatic equations (24),

$$\mathbf{d}(+\infty)=\mathbf{U}_d(+\infty,-\infty)\mathbf{d}(-\infty). \tag{28}$$

This evolution matrix is unitary, and can be parametrized as

$$\mathbf{U}_d(+\infty,-\infty)=\begin{bmatrix}\sqrt{1-p}\,e^{i\xi} & \sqrt{p}\,e^{i\eta}\\ -\sqrt{p}\,e^{-i\eta} & \sqrt{1-p}\,e^{-i\xi}\end{bmatrix}, \tag{29}$$

where p is the probability of nonadiabatic transitions in the effective two-level problem, while ξ and η are dynamical phases; all these depend on the interaction parameters.

For the *counterintuitive* pulse sequence, $\Omega_p(t)=\Omega_2(t)$ and $\Omega_s(t)=\Omega_1(t)$. Then $\vartheta^{ci}(-\infty)=0$ and $\vartheta^{ci}(+\infty)=\pi/2$. The initial conditions (4) can be satisfied only if

$$d_1^{ci}(-\infty)=e^{i\phi},\quad d_2^{ci}(-\infty)=0, \tag{30}$$

where ϕ is an arbitrary unimportant phase. From Eqs. (26)–(30) we find that the populations at $t\to+\infty$ in our three-level system are

$$P_1^{ci}=4p(1-p)\cos^2(\xi+\eta), \tag{31}$$

$$P_2^{ci}=4p(1-p)\sin^2(\xi+\eta), \tag{32}$$

$$P_3^{ci}=(1-2p)^2. \tag{33}$$

For *intuitive* pulses, $\Omega_p(t)=\Omega_1(t)$ and $\Omega_s(t)=\Omega_2(t)$. Then $\vartheta^{i}(-\infty)=\pi/2$ and $\vartheta^{i}(+\infty)=0$. The initial conditions (4) require

$$d_1^{i}(-\infty)=d_2^{i}(-\infty)=\frac{1}{\sqrt{2}}e^{i\phi}, \tag{34}$$

where ϕ is again an unimportant phase. From Eqs. (26)–(29) and (34) we obtain the populations at $t\to+\infty$ to be

$$P_1^{i}=4p(1-p)\cos^2(\xi-\eta), \tag{35}$$

$$P_2^{i}=[(1-p)\sin 2\xi-p\sin 2\eta]^2, \tag{36}$$

$$P_3^{i}=[(1-p)\cos 2\xi-p\cos 2\eta]^2. \tag{37}$$

We should point out again that the parameters p, ξ, and η in Eqs. (31)–(33) and (35–37) are *the same*, as long as the pulse order is reversed by interchanging Ω_1 and Ω_2. This parametrization of the populations leads to several important conclusions.

(i) When the adiabaticity parameter α (the pulse area) increases, the probability p for nonadiabatic transitions in the effective two-level system tends to zero, while ξ is nearly proportional to the adiabatic phase (18), i.e.,

$$p\approx 0,\quad \xi\approx-\tfrac{1}{2}\int_{-\infty}^{\infty}\Omega_0(t)dt=-\tfrac{1}{2}\zeta\quad(\alpha\to\infty).$$

Then $P_3^{ci}\approx 1$ and $P_3^{i}\approx\cos^2\zeta$, i.e., *the final-state population tends to unity for counterintuitive pulses while it oscillates for intuitive pulses* (since ζ is proportional to α), in agreement with Eqs. (15) and (19).

(ii) *In the adiabatic limit,the initial-state population vanishes* for both counterintuitive and intuitive pulse orders, $P_1^{ci}\approx 0$, $P_1^{i}\approx 0$, because then $p\approx 0$. For counterintuitive pulses this is obvious but for the intuitive order it is not.

(iii) If $\dot{\vartheta}(t)$ and $\Omega_0(t)$ are *even functions of time* [which will be the case if, e.g., Eqs. (3) hold], it is well known that the symmetry of Eqs. (24) implies that $\eta=0$ or π. Then *the ultimate initial-state population is the same for both pulse orders*, $P_1^{ci}=P_1^{i}=4p(1-p)\cos^2\xi$. We show in the Appendix that this property holds even *for nonzero intermediate-level detuning* $\Delta(t)$, provided $\Delta(t)$ too is an even function of time, e.g., a constant. Equations (31) and (35) show that even if $\dot{\vartheta}(t)$ and $\Omega_0(t)$ are not symmetric, the initial-state population behaves similarly for both pulse orders, as it has the same amplitude but only a different phase of oscillations.

3. An example: An analytic model

We have found an exact analytic solution on resonance $(\Delta=0)$ for pulses defined by

$$\Omega_1(t)=\Omega_0(t)\cos\Theta(t),\quad \Omega_2(t)=\Omega_0(t)\sin\Theta(t), \tag{38}$$

where

$$\Omega_0(t)=\frac{\alpha}{2T}\operatorname{sech}^2\frac{t}{T},\quad \Theta(t)=\frac{\pi}{4}\left(\tanh\frac{t}{T}+1\right). \tag{39}$$

The pulse $\Omega_1(t)$ precedes the pulse $\Omega_2(t)$ and their maxima are separated by a fixed pulse delay of approximately $0.506T$. The only independent parameter α serves as the adiabaticity parameter: the larger α is the stronger the adiabaticity. Applying $\Omega_1(t)$ to the pump transition and $\Omega_2(t)$ to the Stokes transition or vice versa, one can realize both pulse orders. Note that $\vartheta^{ci}(t)=\Theta(t)$ and $\vartheta^{i}(t)=\pi/2-\Theta(t)$.

The populations for counterintuitive pulses $[\Omega_p(t)=\Omega_2(t)$ and $\Omega_s(t)=\Omega_1(t)]$ are

$$P_1^{ci}=\frac{1}{A^2+1}\sin^2\left(\frac{\pi}{2}\sqrt{A^2+1}\right), \tag{40}$$

$$P_2^{ci}=\left(\frac{2A}{A^2+1}\right)^2\sin^4\left(\frac{\pi}{4}\sqrt{A^2+1}\right), \tag{41}$$

$$P_3^{ci}=\left[1-\frac{2}{A^2+1}\sin^2\left(\frac{\pi}{4}\sqrt{A^2+1}\right)\right]^2, \tag{42}$$

while those for intuitive pulses $[\Omega_p(t)=\Omega_1(t)$ and $\Omega_s(t)=\Omega_2(t)]$ are

$$P_1^{i}=\frac{1}{A^2+1}\sin^2\left(\frac{\pi}{2}\sqrt{A^2+1}\right), \tag{43}$$

$$P_2^{i}=\frac{A^2}{A^2+1}\sin^2\left(\frac{\pi}{2}\sqrt{A^2+1}\right), \tag{44}$$

$$P_3^{i}=\cos^2\left(\frac{\pi}{2}\sqrt{A^2+1}\right), \tag{45}$$

where $A=2\alpha/\pi$ is the area of each pulse. The derivation is straightforward, and it is achieved by changing the independent variable from t to $z=\tanh(t/T)$ and going to the adiabatic representation (24) where the two-state equations involve constant coefficients and are easily solved. Equation (42) shows that, at large α, the probability of nontransfer $1-P_3^{ci}$ for counterintuitive pulses decreases as π^2/α^2. This is yet another example of the breakdown of the Dykhne-Davis-Pechukas exponential dependence [14] reported recently [5,11]. On the other hand, Eq. (45) shows that, for intuitive pulses, the final-state population oscillates between

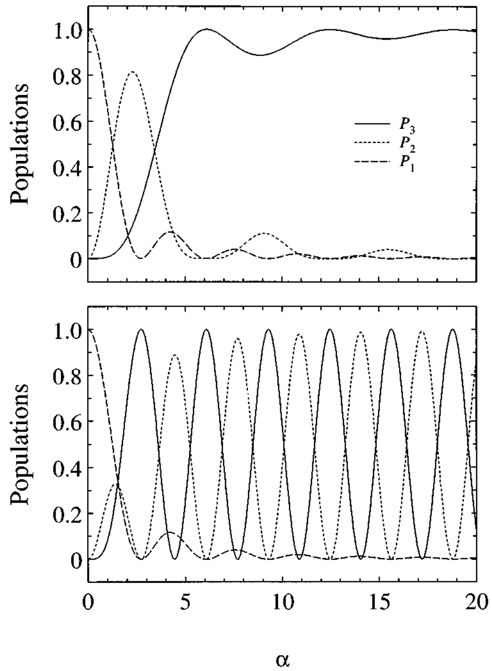

FIG. 4. Populations (40)–(45) for the analytic model (38) plotted against the dimensionless pulse strength α for counterintuitive (upper figure) and intuitive (lower figure) pulse orders.

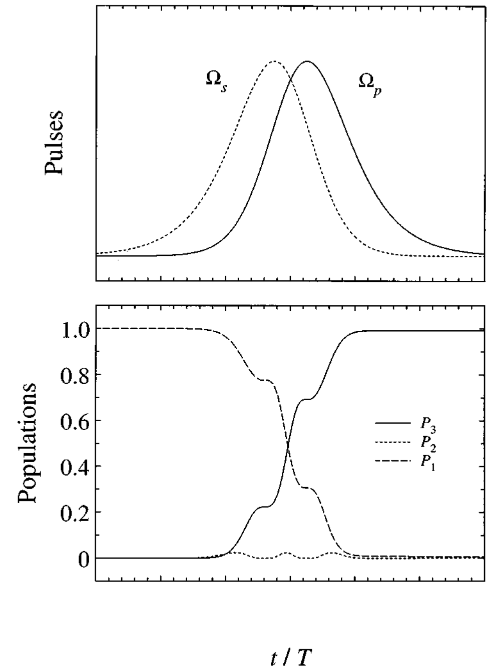

FIG. 5. The pulse shapes and the time evolution of populations (46)–(48) for the analytic model (38) with $\Omega_p=\Omega_2$ and $\Omega_s=\Omega_1$ (counterintuitive pulse order) for $\alpha=20$.

zero and unity when α increases. Note also that the population of level 1 is the same for both intuitive and counterintuitive pulses as for this model, $\Omega_0(t)$ and $\dot{\vartheta}(t)$ are even functions although $\Omega_1(t)$ and $\Omega_2(t)$ are not symmetric themselves. It oscillates and its amplitude decreases in a Lorentzian fashion as α grows. These properties can be seen in Fig. 4, where the populations are plotted against α for counterintuitive (upper figure) and intuitive (lower figure) pulse orders.

A useful feature of this model is that it allows a simple analytic solution not only for the final values of the populations but also for their time evolution. For instance, the time-dependent populations for counterintuitive pulses are exactly given by

$$P_1^{\text{ci}}(t)=\left\{\cos\Theta(t)+\frac{2\sin\chi(t)}{\sqrt{A^2+1}}\left[\cos\chi(t)\sin\Theta(t)-\frac{\sin\chi(t)\cos\Theta(t)}{\sqrt{A^2+1}}\right]\right\}^2, \tag{46}$$

$$P_2^{\text{ci}}(t)=\left(\frac{2A}{A^2+1}\right)^2\sin^4\chi(t), \tag{47}$$

$$P_3^{\text{ci}}(t)=\left\{\sin\Theta(t)-\frac{2\sin\chi(t)}{\sqrt{A^2+1}}\left[\cos\chi(t)\cos\Theta(t)+\frac{\sin\chi(t)\sin\Theta(t)}{\sqrt{A^2+1}}\right]\right\}^2, \tag{48}$$

where $\Theta(t)$ is given by Eq. (39), $\chi(t)=\frac{1}{2}\sqrt{A^2+1}\Theta(t)$ and, as above, $A=2\alpha/\pi$ is the area of each pulse. Evidently, at large α the maximum population of the intermediate state is $4/A^2=\pi^2/\alpha^2\ll 1$. The time evolution of populations (46)–(48) is shown in Fig. 5 for $\alpha=20$ (lower figure), along with the pulse shapes (upper figure). In addition, we can also calculate exactly the probability for nonadiabatic transitions in the effective two-level problem, Eqs. (24) and (30); it is

$$|d_2(t)|^2=\frac{1}{A^2+1}\sin^2\left[\frac{1}{2}\sqrt{A^2+1}\Theta(t)\right], \tag{49}$$

and it vanishes as $1/A^2$.

Equations (46)–(48) are given in a form that shows explicitly the nonadiabatic contributions to the perfectly adiabatic solution $P_{1,\text{ad}}^{\text{ci}}(t)=\cos^2\Theta(t)$, $P_{2,\text{ad}}^{\text{ci}}(t)=0$, and $P_{3,\text{ad}}^{\text{ci}}(t)=\sin^2\Theta(t)$. For instance, we see that for near-adiabatic evolution ($\alpha\gg 1$), the nonadiabatic contribution to $P_3^{\text{ci}}(t)$ is of the order $\mathcal{O}(\alpha^{-1})$, and it introduces (small) oscillations due to the terms with $\chi(t)$ because $\chi(t)$ changes from 0 to $\frac{1}{4}\pi\sqrt{A^2+1}\gg 1$ [in contrast, no oscillations arise from the terms with $\Theta(t)$ as $\Theta(t)$ changes from 0 to $\pi/2$].

Finally, model (38) is unique in the sense that the ratio $|\dot{\vartheta}(t)|/\Omega_0(t)$ is time independent and equals $\pi/(2\alpha)\equiv 1/A$, which implies that the nonadiabatic coupling $\dot{\vartheta}(t)$ vanishes at infinity simultaneously and *in the same manner* as the eigenvalue difference $\Omega_0(t)$. Hence we should not expect appreciable nonadiabatic transitions to take place there, which indeed is seen explicitly in Eq. (49). This fact, along with Eq. (49), suggests that for model (38) there are no particular nonadiabatic regions like, for example, the pulses wings in the case of Gaussian pulses.

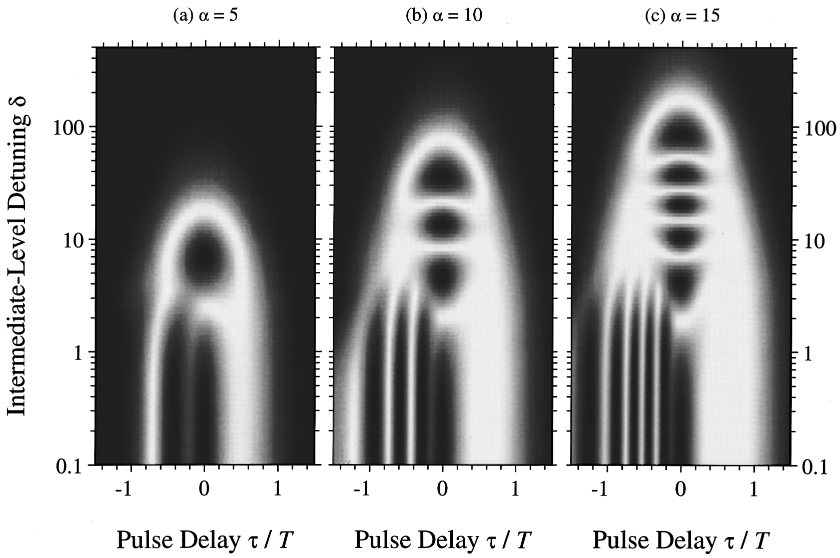

FIG. 6. Intensity plot of the final-state population P_3 for Gaussian pulses, Eq. (53), as a function of the dimensionless pulse delay τ/T and the dimensionless detuning δ for (a) $\alpha=5$, (b) $\alpha=10$, and (c) $\alpha=15$. The white means $P_3=1$, while the black means $P_3=0$.

B. At large detuning

1. Effective two-level problem

It is well known that a N-level system on $(N-1)$-photon resonance can be reduced to an effective two-level system by *adiabatic elimination* of the intermediate levels if the intermediate-level detunings are large compared with the Rabi frequencies. This approximation, which is frequently used in multiphoton absorption, has been discussed in some detail in Refs. [15] and [16]. In this approximation, the three-level system on two-photon resonance is equivalent to a two-level system comprising states $|1\rangle$ and $|3\rangle$ only,

$$i\frac{d}{dt}\begin{bmatrix} c_1(t) \\ c_3(t) \end{bmatrix} \approx \begin{bmatrix} -\Delta_{\rm eff}(t) & \Omega_{\rm eff}(t) \\ \Omega_{\rm eff}(t) & \Delta_{\rm eff}(t) \end{bmatrix} \begin{bmatrix} c_1(t) \\ c_3(t) \end{bmatrix}, \tag{50}$$

given that $\Delta \gg \Omega_{p,s}$. The effective detuning and Rabi frequency are

$$\Delta_{\rm eff} = \frac{\Omega_p^2 - \Omega_s^2}{2\Delta}, \quad \Omega_{\rm eff} = -\frac{\Omega_p \Omega_s}{\Delta}, \tag{51}$$

and the initial conditions are given by Eqs. (4),

$$c_1(-\infty)=1, \quad c_3(-\infty)=0. \tag{52}$$

Equations (50) are obtained from Eqs. (1) when one sets $dc_2(t)/dt=0$, and then solves for $c_2(t)$ from the resulting algebraic equation.

2. Intuitive and counterintuitive pulse orders

Equations (51) show that delayed pulses make the effective detuning $\Delta_{\rm eff}(t)$ chirped. Furthermore, one easily finds that, although $\Delta_{\rm eff}(t)$ vanishes at $t\to\pm\infty$, the ratio $\Delta_{\rm eff}(t)/\Omega_{\rm eff}(t)$ tends to $\pm\infty$ at $t\to-\infty$ and to $\mp\infty$ at $t\to+\infty$ [the upper (lower) sign being for the counterintuitive (intuitive) pulse order], which is a necessary condition for adiabatic inversion. Thus the high efficiency of STIRAP can be viewed as due to a Landau-Zener-type transition in this effective two-level system. We should point out that this argument holds equally well for both pulse orders. Moreover, Eqs. (51) show that reversing the pulse order by interchanging $\Omega_p(t)$ and $\Omega_s(t)$ changes only the sign of $\Delta_{\rm eff}(t)$. It can readily be shown that this leads to the change $c_1(t)\to c_1^*(t)$, $c_3(t)\to -c_3^*(t)$. Thus *the populations for counterintuitive and intuitive pulse orders are approximately the same* at large intermediate-level detuning Δ. The transfer efficiency plotted as a function of the pulse delay τ is generally expected to possess two maxima, one for the intuitive pulse order and another for the counterintuitive pulse order. Indeed, this feature has been observed experimentally [8,4]. Moreover, if Eqs. (3) are chosen, e.g., if the pump and the Stokes pulses have the same shapes, widths, and strengths, and the detuning is constant, then these two maxima will have the same profiles and will be symmetrically placed with respect to $\tau=0$.

It is worth pointing out that as far as the effect of the pulse order is concerned, the difference between the cases on resonance ($\Delta=0$) and at large Δ comes out from the initial conditions in the respective effective two-level problem. On resonance, the initial conditions (30) and (34) for the two pulse orders are different, which is crucial. In contrast, at large Δ, the initial conditions (52) are the same for both pulse orders.

3. An example: Gaussian pulses

To check the above conclusions we have integrated Eq. (1) numerically in the case of Gaussian pulses of the same shapes and strengths but separated by a time delay of 2τ, and for a constant intermediate-level detuning,

$$\Omega_p(t) = \frac{\alpha}{T}\exp\left[-\left(\frac{t-\tau}{T}\right)^2\right],$$

$$\Omega_s(t) = \frac{\alpha}{T}\exp\left[-\left(\frac{t+\tau}{T}\right)^2\right], \quad \Delta(t)=\frac{\delta}{T}, \tag{53}$$

where α, δ, and T are positive parameters, while τ can be positive or negative. A positive τ means a counterintuitive pulse order, while a negative τ means an intuitive order.

Both α and δ are dimensionless, while τ and T have the dimension of time. The intensity plots in Fig. 6 show the final-state population as a function of τ/T and δ for $\alpha=5$, 10, and 15. The figure demonstrates how the regions for the intuitive pulse order, dominated by Rabi oscillations at small detuning ($\delta<3$), become almost identical to those for the counterintuitive order at large detuning. As a result, at large δ the final-state population is nearly symmetric versus τ, as expected, because pulses (53) satisfy the symmetry condition (3); otherwise the profile would be asymmetric. We see that, at large detuning, a large transfer efficiency can be realized with both pulse orders. Furthermore, as α increases (and, thus, adiabaticity improves), the ranges of values of δ and τ/T, over which large transfer efficiency is achieved, increase too. It is worth noting the existence of wide ranges of detunings (e.g., $7<\delta<10$ for $\alpha=10$) over which the transfer efficiency is almost unity, irrespective of the pulse delay unless the latter is very large. Finally, the comparison between the three intensity plots shows that, at small δ, the number of oscillations against τ/T increases with α, which is explained by the conclusions of Sec. IV A 2 above. The plots also demonstrate that for zero delay, $\tau/T=0$, the oscillations versus δ again increase with α. This is explained in Sec. IV C below.

One can easily check that the adiabatic condition for the effective two-level problem (50) generally requires $\alpha^2 \gg \delta,1$. Given the condition of validity of the adiabatic-elimination approximation, $\delta \gg \alpha$, we conclude that adiabaticity for large detuning is achieved when $\alpha^2 \gg \delta \gg \alpha \gg 1$. Evidently, for very large detuning ($\delta \gg \alpha^2$), the transfer efficiency decreases since adiabaticity deteriorates [12].

We have to point out that the conclusions deduced from the adiabatic-elimination approximation appear to be valid in a wider region than the approximation itself ($\delta \gg \alpha,1$). For example, this approximation cannot explain the high transfer efficiency and the symmetry against τ in the final-state population for $\alpha<\delta$, as seen in Fig. 6.

C. Completely overlapping pulses

Consider now the case when the pulses overlap exactly. Let us assume that the pulses have the same time dependence but possibly different strengths,

$$\Omega_p(t)=\frac{\alpha}{T}f\left(\frac{t}{T}\right), \quad \Omega_s(t)=\frac{\beta}{T}f\left(\frac{t}{T}\right), \tag{54}$$

where the parameters α, β, and T are real and positive and $\int_{-\infty}^{\infty}f(x)dx=1$. Evidently, $\vartheta(t)\equiv\text{const}$ [see Eq. (6)], and $\dot{\vartheta}\equiv 0$. This implies that only the matrix elements in the corners of H_a [Eq. (12)] are nonzero and, hence, the adiabatic state $|0\rangle$ is decoupled from the other adiabatic states. Thus the three-level problem (11) is reduced to an effective two-level one for the adiabatic states $|+\rangle$ and $|-\rangle$,

$$i\frac{d}{dt}\begin{bmatrix} a_+ \\ a_- \end{bmatrix}=\begin{bmatrix} \Omega_0\cot\varphi & i\dot{\varphi} \\ -i\dot{\varphi} & -\Omega_0\tan\varphi \end{bmatrix}\begin{bmatrix} a_+ \\ a_- \end{bmatrix}. \tag{55}$$

It can be shown in general that the properties of these equations imply that the population dynamics of the actual (bare) states should be dominated by Rabi oscillations. We will not present the general analysis here as it involves cumbersome, though straightforward, algebra. Instead, it is more instructive to consider explicitly several particular cases.

1. Adiabatic regime

Let us find the solution in the adiabatic regime $[|\dot{\varphi}(t)| \ll \Omega_0(t)\cot\varphi, \Omega_0(t)\tan\varphi]$. In addition to $\Omega_{p,s}(\pm\infty)=0$, which we assume throughout the paper, we also make the natural assumption that $\Delta(t)$ is a nonzero constant. Then $\varphi(\pm\infty)=0$ [see Eq. (7)] and because the adiabatic Hamiltonian is nearly diagonal, simple calculations give

$$P_3 \approx \left(\frac{2\alpha\beta}{\alpha^2+\beta^2}\right)^2 \sin^2\left\{\frac{1}{4}\int_{-\infty}^{\infty}\left[\sqrt{4\Omega_0^2(t)+\Delta^2}-\Delta\right]dt\right\}, \tag{56}$$

with $\Omega_0=\sqrt{\Omega_p^2+\Omega_s^2}$. Apparently, the final-state population P_3 oscillates as a function of α, β, and Δ, and vanishes at very large detuning Δ. These features can be seen in Fig. 6 (for $\tau=0$) where $\alpha=\beta$. It is worth noting that the oscillation amplitude is largest (unity) for equal pulse strengths, $\alpha=\beta$. If we fix one of the pulse strengths, say β, and increase the other (α), then the final-state population decreases as α^{-2} (and in an oscillatory manner), and eventually tends to zero for $\alpha \gg \beta$.

2. Resonance

On resonance, $\Delta=0$, we have $\varphi\equiv\pi/4$ and $\dot{\varphi}\equiv 0$. The coupling between the adiabatic states $|+\rangle$ and $|-\rangle$ vanishes, and the exact solution is easily found

$$P_3=\left(\frac{2\alpha\beta}{\alpha^2+\beta^2}\right)^2 \sin^4\left(\frac{1}{2}\sqrt{\alpha^2+\beta^2}\right). \tag{57}$$

Note that Eq. (57) cannot be obtained from Eq. (56) by setting $\Delta=0$ in the latter, because $\Delta\neq 0$ has been assumed in the derivation of Eq. (56).

3. The same time dependence of the pulses and the detuning

A simple solution is obtained also when the detuning $\Delta(t)$ shares the same time dependence as $\Omega_p(t)$ and $\Omega_s(t)$, that is, when

$$\Delta(t)=\frac{\delta}{T}f\left(\frac{t}{T}\right).$$

Then φ is a (generally nonzero) constant, and $\dot{\varphi}\equiv 0$. The solution is again simple, and the exact final-state population reads

$$P_3=\left(\frac{2\alpha\beta}{\alpha^2+\beta^2}\right)^2\left(\sin^2\varphi\sin^2\frac{\mu+\delta}{4}+\cos^2\varphi\sin^2\frac{\mu-\delta}{4} -\sin^2\varphi\cos^2\varphi\sin^2\frac{\mu}{2}\right), \tag{58}$$

with $\mu=\sqrt{4(\alpha^2+\beta^2)+\delta^2}$. It again oscillates as a function of α, β, and δ. Note that Eq. (58) reduces to Eq. (57) for $\delta=0$, as should be the case.

V. STIRAP AS A LEVEL-CROSSING PROBLEM

A. At large detuning

As we discussed in Sec. IV B, the effective two-level problem (50) at large Δ involves a level crossing, since delayed pulses make the effective detuning $\Delta_{\rm eff}(t)$ chirped. This feature has been noted in Refs. [7] and [13].

B. On resonance

The method of adiabatic elimination, which is applicable for large Δ, and leads to Eq. (50), is completely invalid in the case of intermediate-level resonance, $\Delta=0$. On the other hand, the effective two-level problem for $\Delta=0$, Eqs. (21), does not involve a level crossing. One may then ask if an effective level-crossing two-level problem, that corresponds to STIRAP, exists. We will show that the answer is affirmative, although this two-level problem is quite unusual. Consider the following orthogonal transformation of the probability amplitudes $b_1(t)$ and $b_2(t)$ in Eqs. (21),

$$\mathbf{b}(t)=\mathbf{R}\left(-\frac{\pi}{8}\right)\mathbf{g}(t), \tag{59}$$

where $\mathbf{R}(\phi)$ is the rotation matrix (23). The equations for the amplitudes $\mathbf{g}(t)$ are

$$i\frac{d}{dt}\mathbf{g}(t)=\mathbf{H}_g(t)\mathbf{g}(t), \tag{60}$$

where

$$\mathbf{H}_g(t)=\mathbf{R}\left(\frac{\pi}{8}\right)\mathbf{H}_b(t)\mathbf{R}\left(-\frac{\pi}{8}\right)=\begin{bmatrix}-\Delta_g & \Omega_g\\ \Omega_g & \Delta_g\end{bmatrix},$$

with

$$\Delta_g(t)=\frac{1}{2\sqrt{2}}[\Omega_s(t)-\Omega_p(t)],$$

$$\Omega_g(t)=\frac{1}{2\sqrt{2}}[\Omega_s(t)+\Omega_p(t)]. \tag{61}$$

Obviously, delayed pulses make $\Delta_g(t)$ chirped, and lead to a level crossing at time t_0 where $\Omega_s(t_0)=\Omega_p(t_0)$. From Eqs. (25) and (59) we find that the three-level amplitudes $\mathbf{c}(t)$ are expressed in terms of $\mathbf{g}(t)$ as

$$c_1(t)=\frac{1}{\sqrt{2}}[|g_1(t)|^2-|g_2(t)|^2]-\sqrt{2}\ \mathrm{Re}[g_1(t)g_2^*(t)],$$

$$c_2(t)=-2i\,\mathrm{Im}[g_1(t)g_2^*(t)],$$

$$c_3(t)=\frac{1}{\sqrt{2}}[|g_1(t)|^2-|g_2(t)|^2]+\sqrt{2}\ \mathrm{Re}[g_1(t)g_2^*(t)].$$

The initial conditions (4) require for both pulse orders the same initial conditions for $\mathbf{g}(t)$,

$$g_1(-\infty)=e^{i\phi}\cos\frac{\pi}{8},\quad g_2(-\infty)=-e^{i\phi}\sin\frac{\pi}{8}, \tag{62}$$

where ϕ is an unimportant constant phase. The difference between the two pulse orders, however, still exists. Indeed, reversing the pulse order (by interchanging Ω_p and Ω_s) causes a change of sign in $\Delta_g(t)$ which leads to complex conjugation of the evolution matrix and sign changes in the nondiagonal elements. For initial conditions $\mathbf{g}(-\infty)=(1,0)^T$ or $\mathbf{g}(-\infty)=(0,1)^T$, this does not have any effect, but for the initial conditions (62) it does, of course.

C. The on-resonance chirped two-level problem itself

The chirped two-level problem with detuning and coupling given by Eq. (61), Ω_p and Ω_s being delayed pulses, is interesting by itself, i.e., for the initial conditions

$$g_1(-\infty)=1,\quad g_2(-\infty)=0 \tag{63}$$

rather than Eq. (62), because it demonstrates that *a level crossing and adiabatic evolution do not necessarily imply a transition probability of unity*. Indeed, in the adiabatic limit, the populations are readily found from Eqs. (22), (24) and (59) to be

$$|g_1(+\infty)|^2\approx\tfrac{1}{2}\cos^2\zeta_g,\quad |g_2(+\infty)|^2\approx\tfrac{1}{2}+\tfrac{1}{2}\sin^2\zeta_g,$$

where $\zeta_g=\int_{-\infty}^{\infty}\sqrt{\Omega_g^2(t)+\Delta_g^2(t)}dt$. Thus the ground-state population oscillates between 0 and $\frac{1}{2}$ while the excited-state population oscillates between $\frac{1}{2}$ and 1. The reason for this at a first glance unexpected behavior is that the ratio $\Delta_g(t)/\Omega_g(t)$ does not diverge at $t\to\pm\infty$ (which is a necessary condition for adiabatic inversion), but tends to ∓ 1.

For example, for model (38) with $\Omega_p=\Omega_2$ and $\Omega_s=\Omega_1$, the corresponding Δ_g and Ω_g, Eq. (61), are

$$\Delta_g(t)=-\frac{\alpha}{4T}\mathrm{sech}^2\frac{t}{T}\sin\left(\frac{\pi}{4}\tanh\frac{t}{T}\right),$$

$$\Omega_g(t)=\frac{\alpha}{4T}\mathrm{sech}^2\frac{t}{T}\cos\left(\frac{\pi}{4}\tanh\frac{t}{T}\right). \tag{64}$$

The excited-state population is exactly given by

$$\begin{aligned}|g_2(+\infty)|^2&=1-|g_1(+\infty)|^2\\&=1-\frac{A^2+2}{2(A^2+1)}\cos^2\Bigg(\frac{\pi}{4}\sqrt{A^2+1}\\&\qquad-\frac{1}{2}\arctan\frac{2\sqrt{A^2+1}}{A^2}\Bigg),\end{aligned} \tag{65}$$

where $A=2\alpha/\pi$. The derivation is straightforward, and it is carried out by changing the independent variable from t to $z=\tanh(t/T)$, and going to the adiabatic representation where the two-state equations involve constant coefficients and are easily solved. In Fig. 7, we show the shapes of the pulse and the detuning (64). In Fig. 8, the populations are plotted against α.

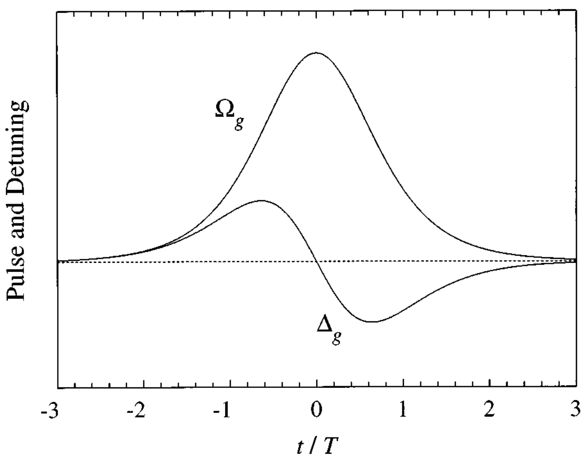

FIG. 7. The shapes of the pulse and the detuning, Eq. (64).

VI. CONCLUSIONS

We have deduced analytically various properties of population transfer by delayed pulses in three-level systems on two-photon resonance in and beyond the adiabatic limit. We have used the fact that the three-level system reduces to effective two-level problems both on resonance ($\Delta=0$) and at large intermediate-level detuning Δ. Special attention was paid to the effect of the pulse order on the population transfer efficiency. We showed that the transfer efficiency depends essentially on the pulse order on resonance, while at large Δ it does not. That is, on resonance, the transfer efficiency approaches steadily unity for the counterintuitive pulse order as the adiabaticity parameter increases, while it oscillates between zero and unity for the intuitive order. At large Δ, both pulse orders produce complete transfer of population to the final state. There is, nevertheless, still a difference between the two pulse orders, since the population transfer is realized via different adiabatic states. Consequently, for the intuitive order the intermediate level is populated during the excitation, while for the counterintuitive order it is not, which is an advantage in the case of strong decay from this state to other states. Furthermore, it is more difficult to achieve adiabatic evolution for the intuitive pulse order. We have also shown that the initial-state population vanishes in the adiabatic limit for both pulse orders. Moreover, under some natural restrictions on the symmetry of the problem, the population of the initial level does not depend on the pulse order for any Δ. We also found that an effective two-level problem exists for completely overlapping pulses as well, which explains why the populations are then dominated by Rabi oscillations. Finally, we demonstrated that STIRAP can be viewed as a level-crossing problem in an equivalent two-level system not only at large Δ (which is known) but also on resonance ($\Delta=0$). The effective on-resonance level-crossing problem is interesting by itself, as it shows that a level crossing and adiabatic evolution do not necessarily lead to complete population inversion. This is another example of the peculiarities of the two-level problems associated with STIRAP in addition to the breakdown of the Dykhne-Davis-Pechukas formula for the probability of nonadiabatic transitions reported recently [5,11].

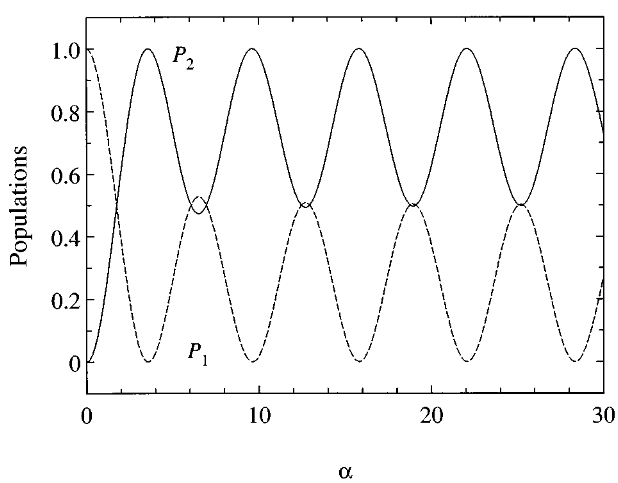

FIG. 8. The populations $P_1=|g_1(+\infty)|^2$ and $P_2=|g_2(+\infty)|^2$, Eq. (65), for the model defined in Eq. (64), plotted against the dimensionless parameter α.

APPENDIX: INDEPENDENCE OF THE INITIAL-STATE POPULATION ON THE PULSE ORDER

We will show that *if* $\dot{\vartheta}(t)$, $\Omega_0(t)$, *and* $\Delta(t)$ *are even functions of time then the ultimate initial-state population is the same for both pulse orders*. For instance, such a case is the frequently considered situation where $\Omega_p(t)$ and $\Omega_s(t)$ have the same symmetric envelopes and the same strengths while the detuning is constant,

$$\Omega_p(t)=\frac{\alpha}{T}f\left(\frac{t-\tau}{T}\right),\quad \Omega_s(t)=\frac{\alpha}{T}f\left(\frac{t+\tau}{T}\right),\quad \Delta(t)=\text{const},$$

and $f(-x)=f(x)$. The proof is an exercise in matrix algebra.

The solution of the adiabatic equations (11) can be expressed in terms of the evolution matrix $\mathbf{U}_a$ as

$$\mathbf{a}(+\infty)=\mathbf{U}_a(+\infty,-\infty)\mathbf{a}(-\infty),$$

where $\mathbf{a}=[a_+,a_0,a_-]^T$. The symmetry of $\Omega_0(t)$ and $\Delta(t)$ means that $\varphi(t)$ is also an even function, $\varphi(-t)=\varphi(t)$ [see Eq. (7)]. Then $\dot{\varphi}(t)$ is an odd function, $\dot{\varphi}(-t)=-\dot{\varphi}(t)$. The implication of the symmetry of the problem is that $\mathbf{U}_a(+\infty,-\infty)$ has the property

$$\mathbf{U}_a^T(+\infty,-\infty)=\mathbf{I}\mathbf{U}_a(+\infty,-\infty)\mathbf{I} \tag{A1}$$

where

$$\mathbf{I}=\begin{bmatrix}1 & 0 & 0\\ 0 & -1 & 0\\ 0 & 0 & 1\end{bmatrix}.$$

In other words, Eq. (A1) implies that

$$(U_a)_{21}=-(U_a)_{12}, \quad (U_a)_{32}=-(U_a)_{23}, \quad (U_a)_{31}=(U_a)_{13}. \tag{A2}$$

To see this, we introduce the evolution matrix $\mathbf{G}(t,0)$, which propagates the system from time $t=0$ to time t,

$$\mathbf{a}(t)=\mathbf{G}(t,0)\mathbf{a}(0).$$

Evidently, the first column of $\mathbf{G}(t,0)$ is the solution of Eqs. (11) for the initial conditions $\mathbf{a}(0)=(1,0,0)^T$, the second column is the solution for the initial conditions $\mathbf{a}(0)=(0,1,0)^T$, and the third column is the solution for the initial conditions $\mathbf{a}(0)=(0,0,1)^T$. We note that time reversal $t\rightarrow -t$ in Eqs. (11) is equivalent to complex conjugation of $\mathbf{a}(t)$ and change of sign of $a_0(t)$. This means that

$$\mathbf{G}(-t,0)=\mathbf{I}\mathbf{G}^*(t,0)\mathbf{I}.$$

Using the unitarity of $\mathbf{U}_a$ and $\mathbf{G}$ and the last equation, we find that

$$\begin{aligned}\mathbf{U}_a(+\infty,-\infty)&=\mathbf{G}(+\infty,0)\mathbf{G}(0,-\infty)=\mathbf{G}(+\infty,0)\mathbf{G}^\dagger(-\infty,0)\\&=\mathbf{G}(+\infty,0)\mathbf{I}\mathbf{G}^T(+\infty,0)\mathbf{I}\end{aligned}$$

and Eq. (A1) follows immediately.

We now return to the basis of the actual (bare) states. Let $\Omega_1(t)$ and $\Omega_2(t)$ be two delayed pulses with $\Omega_1(t)$ preceding $\Omega_2(t)$, Eqs. (27). Suppose first that we have a *counterintuitive* pulse order, in which $\Omega_p(t)=\Omega_2(t)$ and $\Omega_s(t)=\Omega_1(t)$. The probability amplitudes $\mathbf{c}^{\text{ci}}(t)=[c_1^{\text{ci}}(t),c_2^{\text{ci}}(t),c_3^{\text{ci}}(t)]^T$ satisfy the Schrödinger equation (1)

$$i\frac{d}{dt}\mathbf{c}^{\text{ci}}(t)=\mathbf{H}^{\text{ci}}(t)\mathbf{c}^{\text{ci}}(t), \tag{A3}$$

with

$$\mathbf{H}^{\text{ci}}(t)=\begin{bmatrix}0 & \Omega_2(t) & 0\\ \Omega_2(t) & \Delta(t) & \Omega_1(t)\\ 0 & \Omega_1(t) & 0\end{bmatrix}.$$

The population evolution is described by the evolution matrix $\mathbf{U}^{\text{ci}}(t,-\infty)$, and we have

$$\mathbf{c}^{\text{ci}}(+\infty)=\mathbf{U}^{\text{ci}}(+\infty,-\infty)\mathbf{c}^{\text{ci}}(-\infty).$$

Because the bare and adiabatic amplitudes are connected by $\mathbf{c}^{\text{ci}}(t)=\mathbf{W}^{\text{ci}}(t)\mathbf{a}(t)$ [Eq. (9)], we find that

$$\mathbf{U}^{\text{ci}}(+\infty,-\infty)=\mathbf{W}^{\text{ci}}(+\infty)\mathbf{U}_a(+\infty,-\infty)[\mathbf{W}^{\text{ci}}(-\infty)]^\dagger. \tag{A4}$$

Accounting for $\vartheta^{\text{ci}}(-\infty)=0$, $\vartheta^{\text{ci}}(+\infty)=\pi/2$, and $\varphi(-\infty)=\varphi(+\infty)\equiv\varphi$, we find that

$$U_{11}^{\text{ci}}=(U_a)_{12}\sin\varphi+(U_a)_{32}\cos\varphi.$$

Provided the system has initially been in state $|1\rangle$, the population of this state at $t\rightarrow+\infty$ is $P_1^{\text{ci}}=|U_{11}^{\text{ci}}|^2$.

Suppose now that we have an *intuitive* pulse order, in which $\Omega_p(t)=\Omega_1(t)$ and $\Omega_s(t)=\Omega_2(t)$. The probability amplitudes $\mathbf{c}^{\text{i}}(t)=[c_1^{\text{i}}(t),c_2^{\text{i}}(t),c_3^{\text{i}}(t)]^T$ satisfy the Schrödinger equation (1),

$$i\frac{d}{dt}\mathbf{c}^{\text{i}}(t)=\mathbf{H}^{\text{i}}(t)\mathbf{c}^{\text{i}}(t), \tag{A5}$$

with

$$\mathbf{H}^{\text{i}}(t)=\begin{bmatrix}0 & \Omega_1(t) & 0\\ \Omega_1(t) & \Delta(t) & \Omega_2(t)\\ 0 & \Omega_2(t) & 0\end{bmatrix}.$$

The population evolution is described by the evolution matrix $\mathbf{U}^{\text{i}}(t,-\infty)$, and we have

$$\mathbf{c}^{\text{i}}(+\infty)=\mathbf{U}^{\text{i}}(+\infty,-\infty)\mathbf{c}^{\text{i}}(-\infty).$$

Obviously, the Hamiltonians for the two pulse orders are related by

$$\mathbf{H}^{\text{i}}(t)=\mathbf{K}\mathbf{H}^{\text{ci}}(t)\mathbf{K}, \quad \mathbf{K}=\begin{bmatrix}0&0&1\\0&1&0\\1&0&0\end{bmatrix}.$$

Therefore, the transformation $\mathbf{c}^i(t)=\mathbf{K}\tilde{\mathbf{c}}(t)$ casts Eqs. (A5) into equations of the same form as Eqs. (A3). Hence

$$\mathbf{U}^{\text{i}}(+\infty,-\infty)=\mathbf{K}\mathbf{U}^{\text{ci}}(+\infty,-\infty)\mathbf{K}. \tag{A6}$$

Equations (A2), (A4), and (A6) lead to the conclusion that

$$\begin{aligned}U_{11}^{\text{i}}=U_{33}^{\text{ci}}&=-(U_a)_{21}\sin\varphi-(U_a)_{23}\cos\varphi\\&=(U_a)_{12}\sin\varphi+(U_a)_{32}\cos\varphi=U_{11}^{\text{ci}}.\end{aligned}$$

Provided the system has initially been in state $|1\rangle$, the population of this state at $t\rightarrow+\infty$ is $P_1^{\text{i}}=|U_{11}^{\text{i}}|^2=|U_{11}^{\text{ci}}|^2=P_1^{\text{ci}}$. Thus the assumption that $\dot{\vartheta}(t)$, $\Omega_0(t)$, and $\Delta(t)$ are even functions of time led us to the conclusion that the ultimate population of the initial state $|1\rangle$ does not depend on the pulse order.

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