Population transfer by delayed pulses via continuum states

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This paper analyzes the process of population transfer between two discrete states via a common continuum by using two partially overlapping delayed laser pulses. The incoherent ionization channels, the Stark shifts, and the nonzero Fano parameter are taken into account. An analytic approximation to the final-state population is derived for Gaussian pulse shapes. [S1050-2947(97)05307-9]

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I. INTRODUCTION

Stimulated Raman adiabatic passage (STIRAP [1–3], and references therein) is a well established and efficient technique for population transfer between two discrete atomic or molecular states via an intermediate state $|i\rangle$ [Fig. 1(a)]. This process requires three conditions: *two-photon resonance* between the initial (ground) state $|g\rangle$ and the final (excited) state $|e\rangle$, *counterintuitive pulse order*, in which the Stokes pulse, driving the transition between states $|i\rangle$ and $|e\rangle$, precedes the pump pulse, driving the transition between states $|g\rangle$ and $|i\rangle$, though they must overlap partly, and *adiabatic time evolution*. STIRAP exploits the existence of an eigenstate of the Hamiltonian, which involves the bare states $|g\rangle$ and $|e\rangle$ only. In the adiabatic limit, the population is completely transferred to the final state $|g\rangle$ and no population resides in the intermediate state $|i\rangle$ at any time.

It has recently been suggested by Carroll and Hioe [4] that population transfer by a counterintuitive pulse sequence may be realized even if the *discrete* intermediate state [Fig. 1(a)] is replaced by a *continuum* [Fig. 1(b)]. An attractive feature of such a scheme is its flexibility and generality. The Carroll-Hioe analytic model, which involves a quasicontinuum, suggests that complete population transfer is possible, the ionization being suppressed. Later, it was demonstrated by Nakajima *et al.* [5] that this result derives from the very stringent restrictions of the model, which are unlikely to be met in a realistic physical system. These latter authors considered in particular the role of the nonzero Fano parameter q in deteriorating the transfer efficiency. They have also suggested that other loss mechanisms, such as incoherent ionization channels, dynamic Stark shifts, and continuum-continuum transitions, may reduce the transfer efficiency by orders of magnitude; however, this has been disputed recently [6–8].

In the present paper, we study the effect of the incoherent ionization channels, the Stark shifts, and the Fano parameter on the transfer efficiency. We ignore continuum-continuum transitions that are significant for very high laser intensities only. This paper is organized as follows. In Sec. II, we introduce the basic equations and definitions. In Sec. III, we make an analogy between the scheme with a continuum and the standard STIRAP, which provides a better insight into the process. In Sec. IV, we analyze the problem in terms of the adiabatic states and the dark and bright states and derive the adiabatic solution. In Sec. V, we illustrate our conclusions with numerical results for a realistic model. In Sec. VI, we derive an analytic approximation to the final-state population for Gaussian pulse shapes, which describes the effect of the incoherent channels in the case of effective two-photon resonance [7,8]. Finally, in Sec. VII, we present the conclusions.

II. DEFINITION OF THE PROBLEM

The problem of two bound states coupled by two laser fields via a common continuum has been studied intensively in the context of laser-induced continuum structure (LICS [9], and references therein). The time evolution of the probability amplitudes $c_g(t)$ and $c_e(t)$ of the two bound states is governed by the equation [9]

$$i\frac{d}{dt}\begin{bmatrix} c_g \\ c_e \end{bmatrix} = \begin{bmatrix} \Sigma_g - \frac{1}{2}i\Gamma_g & -\frac{1}{2}\sqrt{\Gamma_g^p\Gamma_e^s}(q+i) \\ -\frac{1}{2}\sqrt{\Gamma_g^p\Gamma_e^s}(q+i) & \Sigma_e - \frac{1}{2}i\Gamma_e + D \end{bmatrix} \begin{bmatrix} c_g \\ c_e \end{bmatrix}. \tag{1}$$

Equation (1) is obtained from the Schrödinger equation by adiabatic elimination of the continuum states and within the rotating-wave approximation. The constant q, called the Fano parameter, is responsible for the asymmetric dependence of the ionization on the two-photon detuning D in LICS. It plays an important role in the context of population transfer too [5]. The quantities $\Gamma_g = \Gamma_g^p + \Gamma_g^s$ and $\Gamma_e = \Gamma_e^p + \Gamma_e^s$ are the total ionization widths of states $|g\rangle$ and $|e\rangle$, respectively, which are given by sums of the ionization widths due to the pump and Stokes pulses, whereas $\Sigma_g = \Sigma_g^p + \Sigma_g^s$ and $\Sigma_e = \Sigma_e^p + \Sigma_e^s$ are the corresponding dynamic Stark shifts of states $|g\rangle$ and $|e\rangle$. The ionization widths and the Stark shifts are proportional to the pulse intensities $I_p(t)$ and $I_s(t)$,

$$\Gamma_x^y(t) = G_x^y I_y(t), \quad \Sigma_x^y(t) = S_x^y I_y(t) \quad (x=g,e; y=p,s),$$

where the parameters G_x^y and S_x^y depend on the particular atomic states and the laser frequencies. For the moment we do not impose any restrictions on the shapes of $I_p(t)$ and $I_s(t)$ but only require that they vanish at infinity and their

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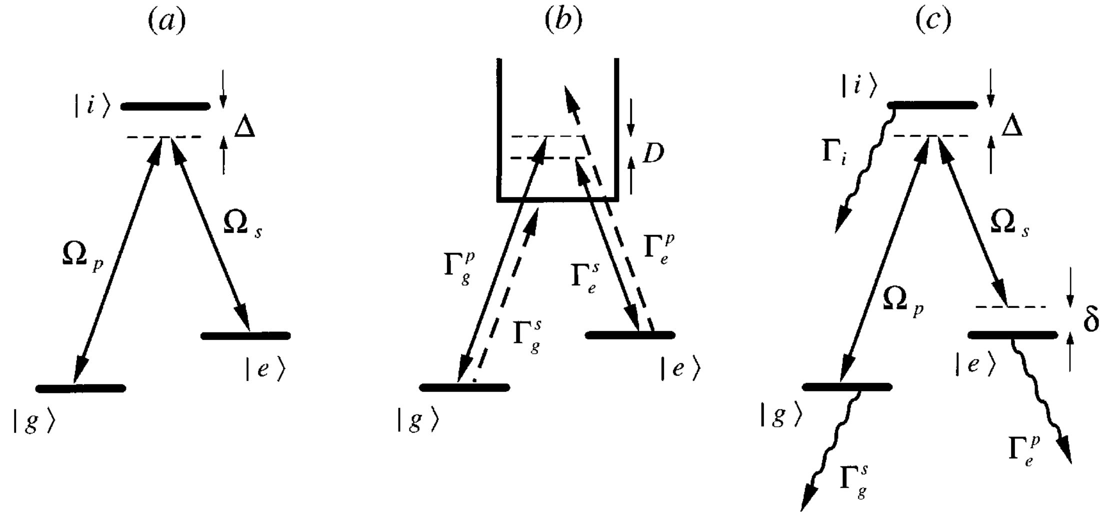

FIG. 1. (a) The three-state Λ system in the standard STIRAP involving a *discrete* intermediate state $|i\rangle$. The initial (ground) state $|g\rangle$ and the final (excited) state $|e\rangle$ are on two-photon resonance while the intermediate state may be detuned by a certain detuning Δ. The Stokes pulse Ω_s precedes the pump pulse Ω_p (counterintuitive pulse order). (b) The population transfer scheme with a *continuum* replacing the discrete intermediate state. The solid arrows indicate the Raman transition between $|g\rangle$ and $|e\rangle$ while the dashed arrows show the incoherent ionization channels. One of the incoherent channels may be avoided (as shown for Γ_g^s) but at least one (Γ_e^p) is always present. The two-photon detuning is denoted by D. (c) The equivalent Λ system corresponding to the continuum scheme in (b). Γ_g^s, Γ_e^p, and Γ_i are the irreversible-decay rates of the three states while δ and Δ are the two-photon detuning and the intermediate-state detuning, respectively. The relations between the quantities in (b) and (c) are given by Eqs. (5).

areas are finite. In the counterintuitive pulse sequence, the Stokes pulse precedes the pump pulse, i.e., $\lim_{t\to-\infty}[I_p(t)/I_s(t)]=0$, $\lim_{t\to+\infty}[I_p(t)/I_s(t)]=\infty$, while in the intuitive sequence the pump pulse comes first. Finally, we assume that at the initial time $t\to-\infty$ the system is in its ground state, $|c_g(-\infty)|=1$, $c_e(-\infty)=0$, and we are interested in the populations at $t\to+\infty$ of the bound states, $P_x=|c_x(+\infty)|^2$ $(x=g,e)$, and the continuum, $P_c=1-P_g-P_e$.

The pump pulse applied on the $|g\rangle$-continuum transition and the Stokes pulse applied on the $|e\rangle$-continuum transition [the solid arrows in Fig. 1(b)] form a two-photon Raman transition that leads to population transfer between states $|g\rangle$ and $|e\rangle$. In contrast, the pump pulse applied on the $|e\rangle$-continuum transition as well as the Stokes pulse applied on the $|g\rangle$-continuum transition [the dashed arrows in Fig. 1(b)] cause irreversible ionization. These two incoherent ionization channels, which have been neglected in earlier studies [4,5], turn out to be the main problem for population transfer. At least one of them is always present [as shown in Fig. 1(b)], which prevents complete population transfer.

III. EQUIVALENT STIRAP PROBLEM

In order to compare the continuum scheme to STIRAP, we notice that by means of the (population preserving) phase transformation

$$c_x(t)=b_x(t)\exp\left\{-i\int_{-\infty}^{t}\left[\Sigma_g(t')+\frac{1}{2}q\Gamma_g^p(t')\right]dt'\right\} \qquad (x=g,e),$$

Eq. (1) takes the form

$$i\frac{d}{dt}\begin{bmatrix} b_g\\ b_e\end{bmatrix} = \begin{bmatrix} -\frac{1}{2}\Gamma_g^p(q+i)-\frac{1}{2}i\Gamma_g^s & -\frac{1}{2}\sqrt{\Gamma_g^p\Gamma_e^s}(q+i)\\ -\frac{1}{2}\sqrt{\Gamma_g^p\Gamma_e^s}(q+i) & -\frac{1}{2}\Gamma_e^s(q+i)-\frac{1}{2}i\Gamma_e^p+\delta\end{bmatrix} \times\begin{bmatrix} b_g\\ b_e\end{bmatrix}, \qquad (2)$$

where

$$\delta=D+\Sigma_e-\Sigma_g-\tfrac{1}{2}q(\Gamma_g^p-\Gamma_e^s). \qquad (3)$$

The last term $\frac{1}{2}q(\Gamma_g^p-\Gamma_e^s)$ in δ is exactly the trapping detuning, which plays an important role in LICS [9]. For $\Gamma_g^s=\Gamma_e^p=0$, Eq. (2) reminds us of the equation

$$i\frac{d}{dt}\begin{bmatrix}\tilde{b}_g\\ \tilde{b}_e\end{bmatrix}=-\frac{1}{\Delta-i\Gamma_i}\begin{bmatrix}\Omega_p^2 & \Omega_p\Omega_s\\ \Omega_p\Omega_s & \Omega_s^2-\delta(\Delta-i\Gamma_i)\end{bmatrix}\begin{bmatrix}\tilde{b}_g\\ \tilde{b}_e\end{bmatrix}. \qquad (4)$$

Equation (4) describes STIRAP in the standard three-state configuration when the intermediate-state detuning Δ and/or rate of decay out of the system Γ_i are large compared to the two-photon detuning δ and the Rabi frequencies Ω_p and Ω_s of the pump and Stokes pulses. It is obtained from the Schrödinger equation by adiabatic elimination of the intermediate state [2,3]. The relations between the variables in Eqs. (2) and (4) are

$$\Gamma_g^p = \frac{2\Omega_p^2\Gamma_i}{\Delta^2+\Gamma_i^2}, \quad \Gamma_e^s = \frac{2\Omega_s^2\Gamma_i}{\Delta^2+\Gamma_i^2}, \quad q = \frac{\Delta}{\Gamma_i}. \tag{5}$$

Hence, the process of population transfer via the continuum [Fig. 1(b)] is equivalent to the standard STIRAP in a three-state system [Fig. 1(a)] but with two-photon detuning δ (induced by the Stark shifts and the Fano parameter) and (time-dependent) irreversible-decay rates Γ_g^s and Γ_e^p of the ground and excited states; this effective three-state system is shown in Fig. 1(c). Thus, the transfer efficiency in the continuum scheme is deteriorated by the incoherent channels, the Stark shifts and the Fano parameter in the same way as the STIRAP efficiency is deteriorated by the irreversible decay of states $|g\rangle$ and $|e\rangle$ and the two-photon detuning.

IV. ADIABATIC BASIS AND DARK-BRIGHT BASIS

A. Adiabatic states and adiabatic solution

Inasmuch as STIRAP gives unity transfer efficiency in the adiabatic limit, our next step is to find the behavior of the transfer efficiency in the continuum scheme as the adiabatic limit is approached. For this we need to go to the adiabatic representation, that is to the basis of the instantaneous eigenstates of the Hamiltonian in Eq. (1). The probability amplitudes $a_-(t)$ and $a_+(t)$ of the adiabatic states $|-\rangle$ and $|+\rangle$ are connected to the bare (diabatic) amplitudes by the transformation

$$\begin{bmatrix} b_g(t) \\ b_e(t) \end{bmatrix} = \begin{bmatrix} \cos\Theta(t) & \sin\Theta(t) \\ -\sin\Theta(t) & \cos\Theta(t) \end{bmatrix} \begin{bmatrix} a_-(t) \\ a_+(t) \end{bmatrix}, \tag{6}$$

where

$$\sin 2\Theta = 2\sqrt{\Gamma_g^p\Gamma_e^s}(1-iq)/\gamma, \quad \cos 2\Theta = (\gamma_e - \gamma_g + 2i\delta)/\gamma, \tag{7}$$

$$\gamma = \sqrt{4\Gamma_g^p\Gamma_e^s(1-iq)^2 + (\gamma_e - \gamma_g + 2i\delta)^2} \quad (\mathrm{Re}\gamma > 0),$$

$$\gamma_g = \Gamma_g^p(1-iq) + \Gamma_g^s, \quad \gamma_e = \Gamma_e^s(1-iq) + \Gamma_e^p.$$

In the adiabatic representation, Eq. (2) becomes

$$\frac{d}{dt}\begin{bmatrix} a_- \\ a_+ \end{bmatrix} = \begin{bmatrix} -\frac{1}{4}(\gamma_g+\gamma_e+2i\delta-\gamma) & -d\Theta/dt \\ d\Theta/dt & -\frac{1}{4}(\gamma_g+\gamma_e+2i\delta+\gamma) \end{bmatrix} \times \begin{bmatrix} a_- \\ a_+ \end{bmatrix}. \tag{8}$$

We assume that $\Gamma_g^s < \Gamma_e^s$ and $\Gamma_e^p < \Gamma_g^p$, i.e., the incoherent channels are weaker than those of the Raman transition. Then it can be shown from Eqs. (6) and (7) that for the counterintuitive pulse sequence, the initial conditions are $|a_-(-\infty)|=1$, $a_+(-\infty)=0$, and the final excited-state population P_e is equal to the probability of remaining in the adiabatic state $|-\rangle$. For the intuitive order, the initial conditions are $|a_+(-\infty)|=1$, $a_-(-\infty)=0$, and P_e is equal to the probability of remaining in state $|+\rangle$. For simplicity, we will assume that the detuning $D=0$; then δ is proportional to the laser intensities, while Θ is independent of them. Thus, the diagonal elements in Eq. (8) are proportional to the pulse intensities, whereas the off-diagonal elements are independent of them and inversely proportional to the pulse widths. Hence, for large pulse areas the evolution in the adiabatic basis is nearly diagonal, i.e., adiabatic. The adiabatic solution for the *counterintuitive* pulse sequence is

$$P_e^{\mathrm{ad}} \approx \exp\left\{-\frac{1}{2}\int_{-\infty}^{\infty}[\Gamma_g^p(t)+\Gamma_g^s(t)+\Gamma_e^s(t)+\Gamma_e^p(t) - \mathrm{Re}\gamma(t)]dt\right\}, \tag{9}$$

while for the *intuitive* sequence it is

$$P_e^{\mathrm{ad}} \approx \exp\left\{-\frac{1}{2}\int_{-\infty}^{\infty}[\Gamma_g^p(t)+\Gamma_g^s(t)+\Gamma_e^s(t)+\Gamma_e^p(t) + \mathrm{Re}\gamma(t)]dt\right\}, \tag{10}$$

where $\mathrm{Re}\gamma = \sqrt{\frac{1}{2}[|\gamma^2| + \mathrm{Re}(\gamma^2)]}$. If the ionization widths of the incoherent channels are small compared to the ones of the Raman transition ($\Gamma_g^s, \Gamma_e^p \ll \Gamma_g^p, \Gamma_e^s$), then

$$\gamma \approx (\Gamma_g^p + \Gamma_e^s)(1-iq) + \frac{\Gamma_g^p - \Gamma_e^s}{\Gamma_g^p + \Gamma_e^s}(\Gamma_g^s - \Gamma_e^p - 2i\delta) + \cdots.$$

In the absence of incoherent ionization ($\Gamma_g^s = \Gamma_e^p = 0$) and for effective two-photon resonance ($\delta=0$), the adiabatic solution gives $P_e^{\mathrm{ad}} \approx 1$ for counterintuitive pulses while $P_e^{\mathrm{ad}} \approx \exp\{-\int_{-\infty}^{\infty}[\Gamma_g^p(t)+\Gamma_e^s(t)]dt\}$ for the intuitive order. In other words, in the adiabatic regime the transfer efficiency approaches unity for the *counterintuitive* order, as in the standard STIRAP. This confirms earlier conclusions for the case when the incoherent ionization channels, the Stark shifts, and the Fano parameter are all equal to zero [4–7]. The condition $\delta=0$ can be achieved by using the Stark shifts induced by an additional laser (whose frequency is small enough not to influence the system otherwise), as proposed in [7], or by using appropriately chirped laser pulses [8]. The presence of at least one of the incoherent channels Γ_g^s or Γ_e^p, however, is unavoidable and leads to ionization losses. Moreover, then the integral in Eq. (9) is nonzero and since all terms in the integrand are proportional to the pulse intensities, the transfer efficiency decreases exponentially for large pulse areas. Finally, for the *intuitive* pulse order P_e always vanishes in the adiabatic regime (even for $\Gamma_g^s = \Gamma_e^p = \delta = 0$), unlike STIRAP where efficient population transfer is possible for large intermediate-state detuning and negligible decay rates [2,3].

B. Dark and bright states

Another useful basis is that of the dark and bright states,

$$|\mathrm{dark}\rangle = \cos\vartheta|g\rangle - \sin\vartheta|e\rangle, \quad |\mathrm{bright}\rangle = \sin\vartheta|g\rangle + \cos\vartheta|e\rangle,$$

with $\tan\vartheta(t) = \sqrt{\Gamma_g^p(t)/\Gamma_e^s(t)}$. In the absence of incoherent channels ($\Gamma_g^s = \Gamma_e^p = 0$), and for $\delta = 0$, the dark and bright states coincide with the adiabatic states: $|-\rangle = |\text{dark}\rangle$, $|+\rangle = |\text{bright}\rangle$. The reason for the names "bright" and "dark" is that, as follows from Eq. (1), the total ionization rate is

$$\frac{d}{dt}P_c = \frac{d}{dt}(1 - |c_g|^2 - |c_e|^2)$$
$$= \Gamma_g^s |c_g|^2 + \Gamma_e^p |c_e|^2 + \sqrt{\Gamma_g^p + \Gamma_e^s} |a_{\text{bright}}|^2. \tag{11}$$

Hence, in the absence of incoherent ionization ($\Gamma_g^s = \Gamma_e^p = 0$), only the population in the bright state is exposed to ionization, whereas the ionization of the dark state is suppressed. For the counterintuitive pulse order the dark state coincides with the ground state $|g\rangle$ before the interaction and with state $|e\rangle$ after it, so that complete population transfer is possible (in principle) if the evolution is adiabatic. In contrast, for the intuitive sequence the system is initially in the bright state, and the adiabatic evolution leads to optimal ionization rather than to population transfer to the excited state.

When the incoherent channels are present and/or for nonzero effective two-photon detuning ($\delta \neq 0$), the dark and bright states are no longer adiabatic states; i.e., staying in one of the adiabatic states does not mean staying in the dark state any more. From another point of view, the equations for the amplitudes of the dark and bright states,

$$i\frac{d}{dt}\begin{bmatrix} a_{\text{dark}} \\ a_{\text{bright}} \end{bmatrix} = \begin{bmatrix} u & w - i\dot{\vartheta} \\ w + i\dot{\vartheta} & v \end{bmatrix} \begin{bmatrix} a_{\text{dark}} \\ a_{\text{bright}} \end{bmatrix}, \tag{12}$$

with

$$u = \left(\delta - \frac{1}{2}i\Gamma_e^p\right)\sin^2\vartheta - \frac{1}{2}i\Gamma_g^s\cos^2\vartheta,$$

$$v = \left(\delta - \frac{1}{2}i\Gamma_e^p\right)\cos^2\vartheta - \frac{1}{2}i\Gamma_g^s\sin^2\vartheta - \frac{1}{2}(q+i)(\Gamma_g^p + \Gamma_e^s),$$

$$w = \frac{1}{2}\left[\frac{i}{2}(\Gamma_g^s - \Gamma_e^p) - \delta\right]\sin 2\vartheta,$$

show that the incoherent channels and the effective detuning δ introduce an additional coupling w between the dark and bright states. The incoherent channels induce also irreversible decay of the dark state with a rate $-\text{Im}u$. Thus, even if the system is somehow forced to stay in the dark state, the ionization losses are unavoidable for nonzero Γ_g^s or/and Γ_e^p.

V. NUMERICAL RESULTS

We have integrated Eq. (1) numerically in the case of Gaussian pulses that have the same widths $2T$ and are separated by a time delay of 2τ. For the ionization widths and the Stark shifts we have taken

$$\Gamma_g^p(t) = Af_p(t), \quad \Gamma_g^s(t) = 0,$$
$$\Gamma_e^p(t) = RAf_p(t), \quad \Gamma_e^s(t) = Af_s(t), \tag{13}$$

$$\Sigma_g^p(t) = Af_p(t), \quad \Sigma_g^s(t) = -Af_s(t),$$
$$\Sigma_e^p(t) = Af_p(t), \quad \Sigma_e^s(t) = 3Af_s(t), \tag{14}$$

with

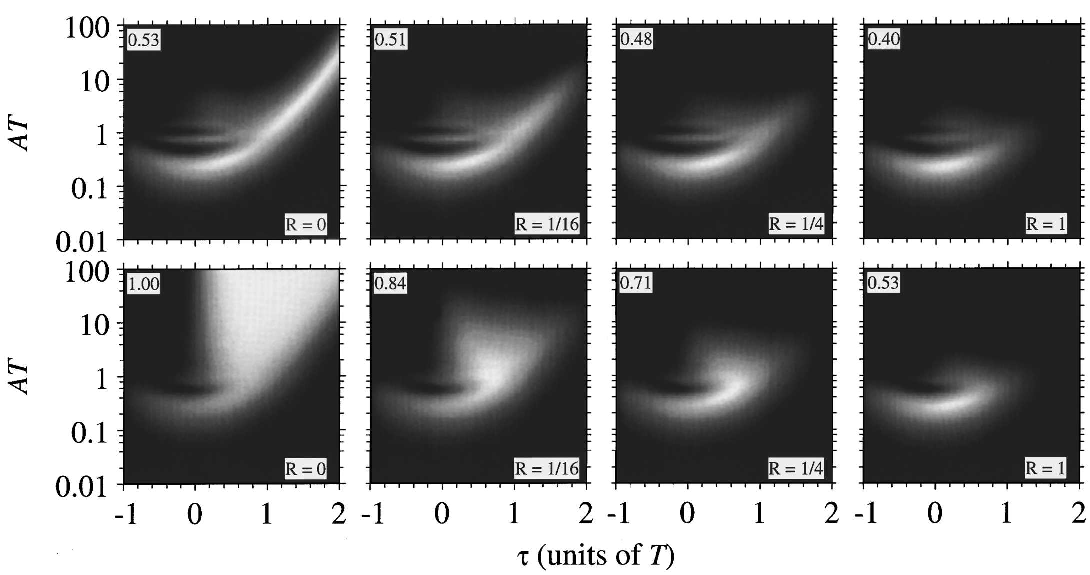

FIG. 2. The excited-state population for Gaussian pulses as a function of the pulse delay τ and AT (essentially the pulse area). Zero is shown by the black while the white displays the maximal population in the particular case (given by the number in the top left corner of each figure). The ionization widths are given by Eqs. (13), the Stark shifts by Eqs. (14), and $q = -6$. Bottom row, effective two-photon resonance ($\delta = 0$); top row, nonzero δ calculated from Eq. (3) with $D = 0$. In the four columns from left to right, we have $R = 0$, $\frac{1}{16}$, $\frac{1}{4}$, and 1 (no, "small," "medium," and "large" incoherent ionization).

$$f_p(t)=\exp\left[-\left(\frac{t-\tau}{T}\right)^2\right],\quad f_s(t)=\exp\left[-\left(\frac{t+\tau}{T}\right)^2\right].$$

The Fano parameter is chosen to be $q=-6$. We have therefore assumed equal pulse areas of Γ_g^p and Γ_e^s and only one incoherent channel present, Γ_e^p. The parameter A is proportional to the laser intensities and the parameter R measures the ratio of the incoherent width Γ_e^p to Γ_g^p. For $R=1/16$, we are very close to the case of population transfer between the $2s^1S_0$ and $5s^1S_0$ states in hydrogen with laser wavelengths $\lambda_p=308.4$ nm and $\lambda_s=1064$ nm [7]. In Fig. 2, we display the population of the excited state $|e\rangle$ as a function of AT (essentially the pulse area) and the time delay τ, for $R=0$ (no incoherent ionization), $R=1/16$ (''small'' incoherent ionization), $R=1/4$ (''medium'' incoherent ionization), and $R=1$ (''large'' incoherent ionization). The results are obtained by numerical integration of Eq. (2). In the lower figures, we have set $\delta=0$ (effective two-photon resonance), whereas in the upper figures we have calculated δ from Eq. (3) with $D=0$. The bottom left figure shows that in the case of effective two-photon resonance and no incoherent channels, the transfer efficiency for $\tau>0$ (counterintuitive pulse order) behaves in the same manner as in STIRAP [2] and exhibits a broad region of large efficiency approaching unity as $AT\rightarrow\infty$. However, for the intuitive pulse order ($\tau<0$), the transfer efficiency is almost zero, unlike STIRAP. Figure 2 also demonstrates that when the incoherent channel Γ_e^p is included, the transfer efficiency decreases considerably and for large incoherent ionization, the continuum scheme is inefficient. We should expect even further decrease in P_e if the other incoherent channel Γ_g^s was open too. The effective two-photon resonance is seen to improve the transfer to some extent, as expected. Slight further improvement can be obtained by optimizing the ratio between the maxima of Γ_g^p and Γ_e^s (chosen unity by us). Furthermore, except for the case with $\Gamma_g^s=\Gamma_e^p=\delta=0$, the transfer efficiency ultimately vanishes for large pulse areas, in agreement with our analysis of the adiabatic regime. Hence, in a realistic situation with incoherent channels, there is an *optimal intensity* (for any fixed pulse delay) where the excited-state population is maximal.

VI. ANALYTIC APPROXIMATION

We have derived an analytic approximation to the excited-state population P_e in the case of effective two-photon resonance ($\delta=0$) by using the approach described below. It is based on the assumption that P_e can be written as a product of two factors: one describing the *losses due to ionization* and another describing the *nonadiabatic losses*. For the former factor we have taken the adiabatic solution P_e^{ad} [Eq. (9)], which approximates P_e well at large intensities. To find the nonadiabatic factor, which is expected to dominate at small intensities, we neglect the incoherent channels as well as the terms i in the factors $(q+i)$ in Eqs. (2) (which is justified for large q); this means that we neglect any ionization. Thus, we obtain a coherent two-state problem that is easily seen (after a phase transformation) to involve a level crossing. In this two-state problem, the effective Rabi frequency and detuning for the Gaussian pulses (13) are (up to an unimportant sign)

$$\Omega_{\mathrm{eff}}(t)=\frac{\xi}{T}e^{-(t/T)^2},\quad \Delta_{\mathrm{eff}}(t)=\frac{\xi}{T}e^{-(t/T)^2}\sinh(2\tau t/T^2),\tag{15}$$

with $\xi=\frac{1}{2}qATe^{-(\tau/T)^2}$. We cannot find analytically the exact solution for these Ω_{eff} and Δ_{eff} but we can approximate it by using some of the available two-state solutions in the manner of Ref. [3]. The most relevant to our problem is the Allen-Eberly model [10],

$$\Omega_{\mathrm{AE}}(t)=\frac{\alpha}{T_0}\mathrm{sech}\frac{t}{T_0},\quad \Delta_{\mathrm{AE}}(t)=\frac{\beta}{T_0}\tanh\frac{t}{T_0}.\tag{16}$$

In order to compensate the differences in the pulse shapes and the detuning chirps in the effective problem (15) and the model (16) as much as possible, we have determined the free parameters α, β and T_0 in such a way that the maxima (at $t=0$) and the areas of $\Omega_{\mathrm{eff}}(t)$ and $\Omega_{\mathrm{AE}}(t)$ as well as the slopes of $\Delta_{\mathrm{eff}}(t)$ and $\Delta_{\mathrm{AE}}(t)$ at the crossing (at $t=0$) are the same. This leads to $\alpha=\xi/\sqrt{\pi}$, $\beta=2\xi\tau/\pi T$, and $T_0=T/\sqrt{\pi}$. The population of state $|e\rangle$ is then given approximately by the transition probability for the model (16),

$$P_e^{\mathrm{AE}}=1-\frac{\cosh^2(\pi\sqrt{\beta^2-\alpha^2})}{\cosh^2(\pi\beta)}.\tag{17}$$

Here, the hyperbolic cosine in the denominator has to be replaced by a cosine for $\alpha>\beta$. We note that in the diabatic basis (2), this is the probability of transition from state $|g\rangle$ to state $|e\rangle$, whereas in the adiabatic basis (8), this is the probability of remaining in state $|-\rangle$.

The excited-state population is approximated as

$$P_e\approx P_e^{\mathrm{ad}}P_e^{\mathrm{AE}}.\tag{18}$$

We point out that the approximation (18) is an intuitive rather than a rigorous result. It gives P_e (which is also the probability of staying in the adiabatic state $|-\rangle$ as the probability P_e^{ad} of nonionization of state $|-\rangle$ times the probability P_e^{AE} of nontransition to the other adiabatic state $|+\rangle$. Since the adiabatic solution (9) gives unity at zero intensity and the correct asymptotics at large intensity, and the solution (17) gives a good approximation for small intensity and unity at infinity, the approximation (18) apparently has the correct limits at both small and large pulse areas; moreover, it turns out to produce fairly good results for moderate pulse areas as well. In Fig. 3, the analytic approximation (18) is compared to the exact values, found by numerical integration of Eq. (1), for various pulse delays τ. We have assumed Gaussian pulse shapes, effective two-photon resonance ($\delta=0$), and we have taken the same case (13) as in Fig. 2, i.e., with the incoherent channel Γ_e^p included. The other parameters are $R=1/16$ and $q=-6$. The approximation (18) is seen to be very precise for small and large intensities and fairly good at moderate intensities, except for $\tau=0.25T$ [11]. It predicts the maximum position and width very well. The small overestimation of P_e is mainly due to the neglect of any ionization mechanism in P_e^{AE}. A better (but more complicated) approximation can be obtained if the ionization widths Γ_g^p and Γ_e^s are accounted for; this is possible and leads to an Allen-Eberly model with complex coefficients that gives the tran-

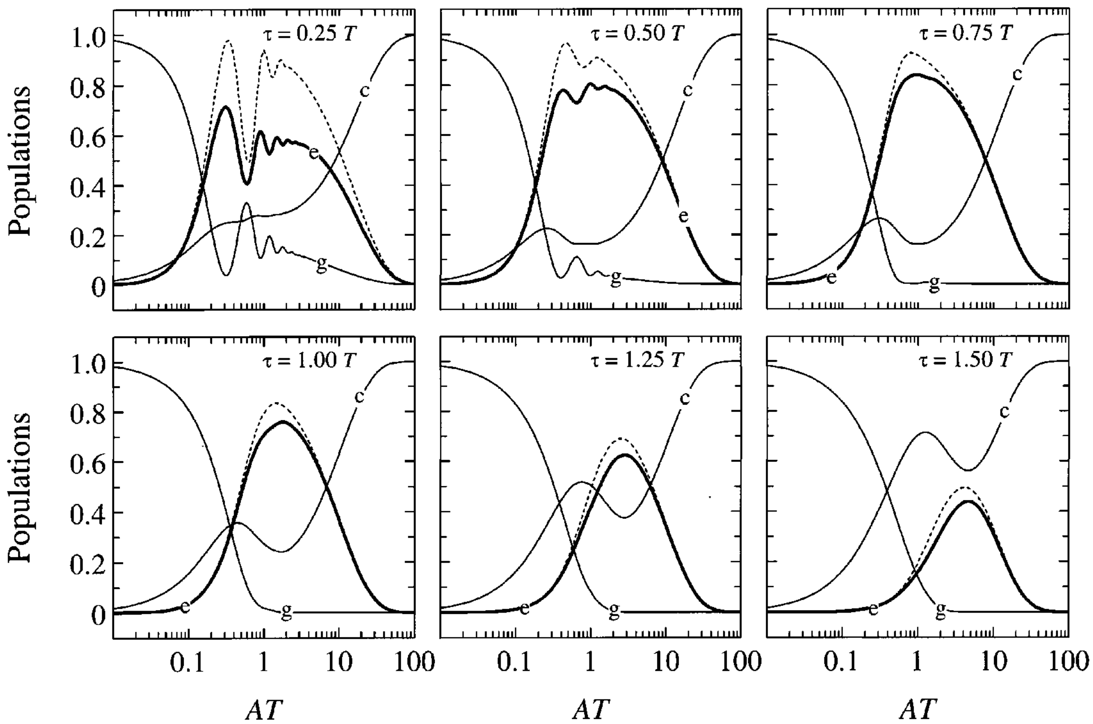

FIG. 3. The populations of the bound states and the continuum for Gaussian pulses against AT (essentially the pulse area) for various pulse delays τ in the case of effective two-photon resonance, $\delta=0$. The solid curves show the exact numerical values and the labels on them refer to the ground state (g), the excited state (e), and the continuum (c). The dashed curves show the analytic approximation (18) for the excited-state population. The ionization widths are given by Eq. (13), $q=-6$ and $R=\frac{1}{16}$.

sition probability in terms of four gamma functions. We also conclude that the analytic solution reproduces to some extent the small oscillations seen around the maximum for small pulse delays. These oscillations are predicted by Eqs. (17) and (18) for $\alpha>\beta$, i.e., for $\tau/T<\sqrt{\pi}/2\approx0.89$. As in the standard STIRAP, these Rabi-like oscillations arise when the overlap between the pulses is appreciable [2]. Finally, Fig. 3 shows that the ground-state population is relatively insensitive to the pulse delay.

VII. CONCLUSIONS

We have investigated analytically and numerically the role of the incoherent ionization channels, the Stark shifts, and the nonzero Fano parameter on the efficiency of the population transfer between two discrete states via a common continuum by means of two partially overlapping delayed laser pulses. At least one incoherent channel is unavoidable and has to be considered in any implementation of such a scheme. We have shown that the incoherent ionization channels correspond to irreversible-decay rates of the initial and final states in the Λ system used in STIRAP, while the Stark shifts and the Fano parameter correspond to a two-photon detuning in STIRAP. We have found that although the transfer efficiency is adversely affected by the incoherent channels, the Stark shifts and the Fano parameter (incomplete) population transfer can still be realized by pulses in the counterintuitive order, while it is virtually impossible by an intuitive pulse sequence. An important difference between the scheme with a continuum and STIRAP is that in the adiabatic limit, the transfer efficiency vanishes in the former scheme while it tends to unity in the latter. There is an optimum range of laser intensities where the transfer efficiency is maximal. We have derived an analytic approximation, Eq. (18), which describes the final-state population in the case when effective two-photon resonance is achieved by Stark shift compensation with an additional laser or by using chirped laser pulses.

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[1] U. Gaubatz, P. Rudecki, M. Becker, S. Schiemann, M. Külz, and K. Bergmann, Chem. Phys. Lett. **149**, 463 (1988); U. Gaubatz, P. Rudecki, S. Schiemann, and K. Bergmann, J. Chem. Phys. **92**, 5363 (1990); B. W. Shore, J. Martin, M. P. Fewell, and K. Bergmann, Phys. Rev. A **52,** 566 (1995); J. Martin, B. W. Shore, and K. Bergmann, *ibid.* **52**, 583 (1995); J. Martin, B. W. Shore, and K. Bergmann, *ibid.* **54**, 1556 (1996); N. V. Vitanov and S. Stenholm, Opt. Commun. **127**, 215 (1996).

[2] N. V. Vitanov and S. Stenholm, Phys. Rev. A **55**, 648 (1997).

[3] N. V. Vitanov and S. Stenholm, Opt. Commun. **135**, 394 (1997).

[4] C. E. Carroll and F. T. Hioe, Phys. Rev. Lett. **68**, 3523 (1992); Phys. Rev. A **47**, 571 (1993); Phys. Lett. A **199**, 145 (1995).

[5] T. Nakajima, M. Elk, J. Zhang, and P. Lambropoulos, Phys. Rev. A **50**, R913 (1994).

[6] C. E. Carroll and F. T. Hioe, Phys. Rev. A **54**, 5147 (1996).

[7] L. P. Yatsenko, R. G. Unanyan, K. Bergmann, T. Halfmann, and B. W. Shore, Opt. Commun. **135**, 406 (1997).

[8] E. Paspalakis, M. Protopapas, and P. L. Knight (unpublished).

[9] P. L. Knight, M. A. Lauder, and B. J. Dalton, Phys. Rep. **190**, 1 (1990).

[10] L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975); F. T. Hioe, Phys. Rev. A **30**, 2100 (1984).

[11] When the pulses overlap too much the population dynamics is increasingly pulse-shape sensitive. This causes inaccuracy because of the differences in the shapes of the actual effective Rabi frequency and detuning (15) and the model ones (16).