Coherent properties of a tripod system coupled via a continuum

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We present results from a study of the coherence properties of a system involving three discrete states coupled to each other by two-photon processes via a common continuum. This tripod linkage is an extension of the standard laser-induced continuum structure (LICS), which involves two discrete states and two lasers. We show that in the tripod scheme, there exist two population trapping conditions; in some cases these conditions are easier to satisfy than the single trapping condition in a two-state LICS. Depending on the pulse timing, various effects can be observed. We derive some basic properties of the tripod scheme, such as the solution for coincident pulses, the behavior of the system in the adiabatic limit for delayed pulses, the conditions for no ionization and for maximal ionization, and the optimal conditions for population transfer between the discrete states via the continuum. In the case when one of the discrete states is strongly coupled to the continuum, the population dynamics reduces to a standard two-state LICS problem (involving the other two states) with modified parameters; this provides the opportunity to customize the parameters of a given two-state LICS system.

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I. INTRODUCTION

Coherent interaction between discrete quantum states via a continuum is an intriguing process. Although the continuum is traditionally seen as an incoherent medium, (partial) transfer of coherence can nevertheless occur through it. In particular, much theoretical and experimental attention has been devoted to the laser-induced continuum structure (LICS) [1–19], where the interaction between a discrete state ψ_2 and a structureless, flat continuum creates a structure in the continuum which significantly affects the interaction of another discrete state ψ_1 with this continuum. For example, the ionization probability for state ψ_1, when plotted as a function of the frequency of the ionizing laser, exhibits the so-called Fano profile [1]. The physical nature of the LICS is closely related to autoionizing states [1,20–26].

It was suggested by Carroll and Hioe a few years ago [27,28] that a continuum can serve as an intermediary for population transfer between two discrete states in an atom or a molecule by using a sequence of two counterintuitively ordered delayed laser pulses. This scheme is an interesting variation of the process of stimulated Raman adiabatic passage (see Refs. [29–32] and references therein) where a discrete intermediate state is used. The Carroll-Hioe analytic model, which involves an infinite quasicontinuum of equidistant discrete states, equally strongly coupled to the two bound states, suggests that complete population transfer is possible, the ionization being suppressed. Later, Nakajima *et al.*, [33] demonstrated that this result derives from the very stringent restrictions of the model which are unlikely to be met in a realistic physical system with a real continuum, in particular with a nonzero Fano parameter q. It has subsequently been recognized that although complete population transfer is unrealistic, significant partial transfer may still be feasible [34–39]. It has been shown that, at least in principle, the detrimental effect of the nonzero Fano parameter and Stark shifts can be overcome by using Stark shifts induced by a third (nonionizing) laser [36] or by using appropriately chirped laser pulses [37,38]. It has been concluded [36,37] that the main difficulty in achieving efficient population transfer is related to the incoherent ionization channels, of which at least one is always present and leads to inevitable irreversible population losses. It has been suggested [35,36] that these losses can be reduced (although not eliminated) by choosing an appropriate region in the continuum where the ionization probability is minimal. Later, it was shown that incoherent ionization can be suppressed very effectively by using a Fano-like resonance induced by an additional laser from a third state ψ_3, resulting in a considerable increase in the transfer efficiency [40].

In the present paper, we investigate the coherence properties of a scheme comprising *three* discrete states coupled via a common continuum. This tripod linkage can be viewed as an extension of the standard LICS, involving two discrete states and two lasers, with the inclusion of an extra state by using a third laser. Such a scheme can also appear in a standard two-state LICS when the two lasers are tuned near an autoionizing state; the latter is strongly coupled to the continuum by configuration interaction. The present scheme can also be viewed as a variation of the tripod scheme comprising three discrete states coupled via a (common) fourth discrete state [41,42]. In contrast to the three-state scheme in Ref. [40], in which the additional laser used to suppress incoherent ionization was tuned in the continuum much above the region where the main lasers are tuned (thus reducing the coupled three-state dynamics to a pair of two-state LICS systems); here the additional laser is tuned in the same region as the two main lasers, which means that we have to deal with

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generally irreducible three-state dynamics. Some properties of this tripod scheme have been studied in Ref. [43] in the particular case when the Fano parameters are equal and the additional state is a strongly coupled autoionizing state. In the present paper we establish the basic properties of this system in the general case of arbitrary Fano parameters and arbitrary strong ionization rates. We derive the population trapping conditions, which are now two, in contrast to the single trapping condition in a two-state LICS. Furthermore, we obtain the solution for coincident pulses and the behavior of the system in the adiabatic limit for delayed pulses, including the optimal conditions for population transfer between the discrete states via the continuum.

This paper is organized as follows. In Sec. II, we introduce the tripod-continuum system, present the basic equations and definitions, and derive the trapping conditions. In Sec. III, we consider the case when all laser fields have the same time dependence. In Sec. IV, we examine the case of delayed laser pulses with a special attention to population transfer in the near-adiabatic regime. In Sec. V, we explore the case when the third state ψ_3 is strongly coupled to the continuum, and eliminate it adiabatically to simplify the dynamics and gain insight into the tripod-continuum interaction. Finally, in Sec. VI we summarize the conclusions.

II. TRIPOD-CONTINUUM SYSTEM

A. System

We shall ignore any continuum-continuum transitions, such as above threshold ionization [44], which become important only for very high laser intensities. We also neglect spontaneous emission from the bound states, which is justified when these states are ground or metastable or when the interaction time is short compared to the atomic relaxation times. Finally, we ignore incoherent ionization channels [36,37,40], i.e., we assume that each laser drives only one transition between a bound state and the continuum.

The total wave function can be written as a linear superposition of the three discrete states and the continuum. We then substitute this expansion into the time-dependent Schrödinger equation and eliminate the continuum using the rotating-wave and Markov approximations [3]. The probability amplitudes of the three bound states are then found to obey the equation ($\hbar=1$)

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

where $\mathbf{C}(t)=[C_1(t),C_2(t),C_3(t)]^T$. The time-dependent Hamiltonian describing the system separates into real and imaginary parts,

$$\mathsf{H}(t)=\mathsf{A}(t)+i\mathsf{B}(t), \tag{2a}$$

$$\mathsf{A}=-\frac{1}{2}\begin{bmatrix} -2\Delta_1 & \sqrt{\Gamma_1\Gamma_2}q_{12} & \sqrt{\Gamma_1\Gamma_3}q_{13} \\ \sqrt{\Gamma_1\Gamma_2}q_{12} & -2\Delta_2 & \sqrt{\Gamma_2\Gamma_3}q_{23} \\ \sqrt{\Gamma_1\Gamma_3}q_{13} & \sqrt{\Gamma_2\Gamma_3}q_{23} & 0 \end{bmatrix}, \tag{2b}$$

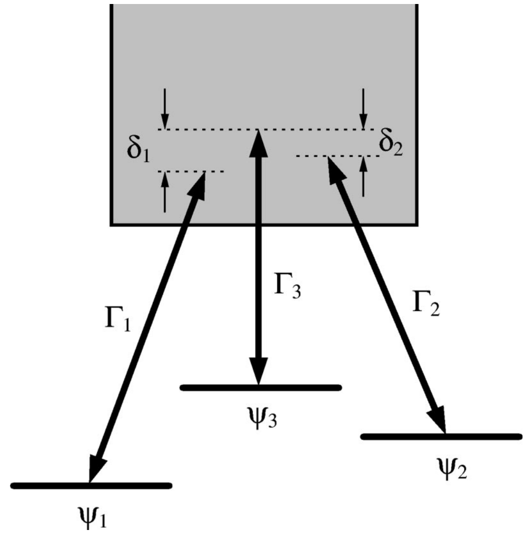

FIG. 1. Sketch of the tripod scheme involving three discrete states ψ_1, ψ_2, and ψ_3 coupled via a common continuum by three lasers. The ionization rates $\Gamma_k(t)$ are proportional to the corresponding laser intensities, and are generally time dependent.

$$\mathsf{B}=-\frac{1}{2}\begin{bmatrix} \Gamma_1 & \sqrt{\Gamma_1\Gamma_2} & \sqrt{\Gamma_1\Gamma_3} \\ \sqrt{\Gamma_1\Gamma_2} & \Gamma_2 & \sqrt{\Gamma_2\Gamma_3} \\ \sqrt{\Gamma_1\Gamma_3} & \sqrt{\Gamma_2\Gamma_3} & \Gamma_3 \end{bmatrix}. \tag{2c}$$

(For typographic simplicity, here and subsequently we often omit the explicit time argument). Here

$$\Delta_1(t)=\delta_1+S_1(t)-S_3(t), \tag{3a}$$

$$\Delta_2(t)=\delta_2+S_2(t)-S_3(t), \tag{3b}$$

with $\delta_k(k=1,2)$ being the static two-photon laser detuning between state ψ_k and state ψ_3,

$$\delta_k=E_k+\omega_k-E_3-\omega_3, \tag{4}$$

where E_k is the energy of state ψ_k and ω_k is the carrier frequency of the laser that couples this state to the continuum. As evident from Eq. (2b) and as shown in Fig. 1, we have chosen the Stark-shifted energy of state ψ_3 as the zero energy level.

The quantity $\Gamma_k(t)(k=1,2,3)$ is the ionization rate of state ψ_k, which is proportional to the generally time-dependent (e.g., pulse-shaped) intensity of the corresponding laser,

$$\Gamma_k(t)=2\pi|V_{k\epsilon}(t)|^2_{\epsilon=E_k+\omega_k}, \tag{5}$$

where $V_{k\epsilon}(t)(k=1,2,3)$ is the interaction operator matrix element between state ψ_k and the continuum state with energy ϵ. $S_k(t)$ $(k=1,2,3)$ is the total laser-induced dynamic Stark shift for state ψ_k, which is a sum of the Stark shifts, induced by each laser. For each laser the Stark shift is proportional to the corresponding laser intensity, and has the form

$$S_k(t)=\mathcal{P}\sum\!\!\!\!\!\!\int_{\epsilon} \frac{|V_{k\epsilon}(t)|^2}{\epsilon-E_k-\omega_k}, \tag{6}$$

where $\mathcal{P}$ is the principal value and the above expression involves summation over all participating bound states and integration over the continuum states.

The dimensionless constants q_{12}, q_{13}, and q_{23} in Eq. (2b) are the Fano asymmetry parameters [1–3,45], which characterize the transitions between the corresponding pairs of states via the continuum and depend on the atomic structure. They are defined by the ratio

$$q_{kl}=\frac{\mathcal{P}\sum\!\!\!\!\!\!\int_{\epsilon} \frac{V_{k\epsilon}(t)V^*_{l\epsilon}(t)}{\epsilon-E_k-\omega_k}}{\pi V_{k\epsilon}(t)V^*_{l\epsilon}(t)}. \tag{7}$$

With the exception of the Fano parameters, all variables involved in Eqs. (2) can be controlled externally by the laser fields and are generally time dependent.

We shall assume that the system is initially in state ψ_1,

$$C_1(-\infty)=1, \quad C_2(-\infty)=C_3(-\infty)=0, \tag{8}$$

and the quantities of interest are the populations of the discrete states at $t\to+\infty$, $P_k=|C_k(+\infty)|^2$ $(k=1,2,3)$, and the ionization probability $P_i=1-P_1-P_2-P_3$. Because we choose the initial conditions (8) and we intend to explore how the additional state ψ_3 affects the interaction between states ψ_1 and ψ_2, we shall refer to $\Gamma_1(t)$, $\Gamma_2(t)$, and $\Gamma_3(t)$ as ionization rates induced by the pump, Stokes, and control lasers, respectively.

B. Eigenvalues and trapping conditions

It was shown in Ref. [46] that if the matrices $\mathsf{A}(t)$ and $\mathsf{B}(t)$ commute,

$$\mathsf{A}(t)\mathsf{B}(t)=\mathsf{B}(t)\mathsf{A}(t), \tag{9}$$

then the eigenvalues of $\mathsf{H}(t)$ read as

$$\lambda_k(t)=\lambda_k^A(t)+i\lambda_k^B(t) \quad (k=1,2,3), \tag{10}$$

where $\lambda_k^A(t)$ and $\lambda_k^B(t)$ are eigenvalues of $\mathsf{A}(t)$ and $\mathsf{B}(t)$, respectively. The importance of relation (10) derives from the fact that the eigenvalues of $\mathsf{B}(t)$ are given by

$$\lambda_1^B(t)=\lambda_2^B(t)=0, \quad \lambda_3^B(t)=-\frac{1}{2}\Gamma(t), \tag{11}$$

where

$$\Gamma(t)=\Gamma_1(t)+\Gamma_2(t)+\Gamma_3(t), \tag{12}$$

i.e., $\mathsf{B}(t)$ has two zero eigenvalues which correspond to *nondecaying* eigenstates of $\mathsf{H}(t)$. The fulfillment of relation (9) requires that

$$\Delta_1(t)=\frac{1}{2}q_{13}[\Gamma_3(t)-\Gamma_1(t)]+\frac{1}{2}(q_{12}-q_{23})\Gamma_2(t), \tag{13a}$$

$$\Delta_2(t)=\frac{1}{2}q_{23}[\Gamma_3(t)-\Gamma_2(t)]+\frac{1}{2}(q_{12}-q_{13})\Gamma_1(t). \tag{13b}$$

Equations (13) will be referred to as *the population trapping conditions*. Hence there are two such conditions imposed on the interaction parameters, rather than just one as in a two-state LICS. It is easily verified that for $\Gamma_3=0$, Eqs. (13) reduce to the well-known trapping condition in a LICS [3]:

$$\Delta_1(t)-\Delta_2(t)=\frac{1}{2}q_{12}[\Gamma_2(t)-\Gamma_1(t)]. \tag{14}$$

Given Eqs. (13), the eigenvalues of $\mathsf{A}(t)$ are

$$\lambda_1^A(t)=a(t)+\sqrt{a^2(t)+b(t)}, \tag{15a}$$

$$\lambda_2^A(t)=a(t)-\sqrt{a^2(t)+b(t)}, \tag{15b}$$

$$\lambda_3^A(t)=-\frac{1}{2}[q_{13}\Gamma_1(t)+q_{23}\Gamma_2(t)], \tag{15c}$$

with

$$a=\frac{1}{4}[q_{13}(\Gamma_3-\Gamma_1)+q_{23}(\Gamma_3-\Gamma_2)+q_{12}(\Gamma_1+\Gamma_2)],$$

$$b=\frac{1}{4}\Gamma_3[q_{13}(q_{13}-q_{12})\Gamma_1+q_{23}(q_{23}-q_{12})\Gamma_2-q_{13}q_{23}\Gamma_3].$$

C. Eigenstates and adiabatic basis

Important information of the interaction dynamics is contained in the instantaneous eigenstates of $\mathsf{H}(t)$—the *adiabatic states*. They are derived readily when the trapping conditions (13) are fulfilled, which we shall assume.

Because of the degeneracy of the two zero eigenvalues of $\mathsf{B}(t)$, there is an ambiguity in the corresponding two eigenstates of $\mathsf{B}(t)$, since any linear combination of them would be a zero-eigenvalue eigenstate of $\mathsf{B}(t)$ too. This implies, in particular, that despite the commutation relation (9), the zero-eigenvalue eigenstates of $\mathsf{B}(t)$ are not necessarily eigenstates of $\mathsf{A}(t)$. Any eigenstate of $\mathsf{A}(t)$, however, is an eigenstate of $\mathsf{B}(t)$, and hence of $\mathsf{H}(t)$ too. The common (time-dependent) eigenstates of $\mathsf{A}(t)$, $\mathsf{B}(t)$, and $\mathsf{H}(t)$ are given by

$$\varphi_1=\begin{bmatrix}\cos\theta\cos\chi-\sin\theta\sin\phi\sin\chi\\ -\sin\theta\cos\chi-\cos\theta\sin\phi\sin\chi\\ \cos\phi\sin\chi\end{bmatrix}, \tag{16a}$$

$$\varphi_2=\begin{bmatrix}\cos\theta\sin\chi+\sin\theta\sin\phi\cos\chi\\ -\sin\theta\sin\chi+\cos\theta\sin\phi\cos\chi\\ -\cos\phi\cos\chi\end{bmatrix}, \tag{16b}$$

$$\varphi_3=\begin{bmatrix}\sin\theta\cos\phi\\ \cos\theta\cos\phi\\ \sin\phi\end{bmatrix}, \tag{16c}$$

where the time-dependent angles $\theta(t)$, $\phi(t)$, and $\chi(t)$ are defined by

$$\tan\theta=\sqrt{\frac{\Gamma_1}{\Gamma_2}}, \tag{17a}$$

$$\tan\phi=\sqrt{\frac{\Gamma_3}{\Gamma_1+\Gamma_2}}, \tag{17b}$$

$$\cot 2\chi=\frac{(\Gamma_1-\Gamma_2)(\Gamma_1+\Gamma_2+2\Gamma_3)}{4\sqrt{\Gamma_1\Gamma_2\Gamma_3(\Gamma_1+\Gamma_2+\Gamma_3)}}+\frac{(\Gamma_1+\Gamma_2)^2(q_{13}+q_{23}-2q_{12})}{4\sqrt{\Gamma_1\Gamma_2\Gamma_3(\Gamma_1+\Gamma_2+\Gamma_3)}(q_{13}-q_{23})}. \tag{17c}$$

The use of adiabatic states is appropriate in two cases—in the near-adiabatic regime and for coincident pulses—because then the couplings between the adiabatic states vanish and it is possible to derive analytic estimates for the population dynamics. We shall do this in Secs. III and IV.

D. Basis of $\varphi_1'(t)$, $\varphi_2'(t)$, and $\varphi_3(t)$

In some cases it is convenient to employ an alternative time-dependent basis composed of states $\varphi_1'(t)$, $\varphi_2'(t)$, and $\varphi_3(t)$, where

$$\varphi_1'=\begin{bmatrix}\cos\theta\\ -\sin\theta\\ 0\end{bmatrix},\quad \varphi_2'=\begin{bmatrix}\sin\theta\sin\phi\\ \cos\theta\sin\phi\\ -\cos\phi\end{bmatrix}, \tag{18}$$

and $\varphi_3(t)$ is the adiabatic state [Eq. (16c)]. Obviously, the adiabatic states $\varphi_1(t)$ and $\varphi_2(t)$ are linear superpositions of states $\varphi_1'(t)$ and $\varphi_2'(t)$:

$$\varphi_1=\varphi_1'\cos\chi-\varphi_2'\sin\chi, \tag{19a}$$

$$\varphi_2=\varphi_1'\sin\chi+\varphi_2'\cos\chi. \tag{19b}$$

Like states $\varphi_1(t)$ and $\varphi_2(t)$, states $\varphi_1'(t)$ and $\varphi_2'(t)$ do not decay; the only decaying state in the $(\varphi_1',\varphi_2',\varphi_3)$ basis is $\varphi_3(t)$. States $\varphi_1'(t)$ and $\varphi_2'(t)$ are (zero-eigenvalue) eigenstates of $\mathsf{B}(t)$, but not generally of $\mathsf{A}(t)$ and $\mathsf{H}(t)$. It can easily be shown that they become eigenstates of $\mathsf{A}(t)$ and $\mathsf{H}(t)$ only when $q_{13}=q_{23}$.

The transformation from the bare-state basis (1) to the $(\varphi_1',\varphi_2',\varphi_3)$ basis, $\mathbf{C}(t)=\mathsf{R}(t)\mathbf{C}'(t)$, is carried out by the time-dependent rotation matrix

$$\mathsf{R}=\begin{bmatrix}\cos\theta & \sin\theta\sin\phi & \sin\theta\cos\phi\\ -\sin\theta & \cos\theta\sin\phi & \cos\theta\cos\phi\\ 0 & -\cos\phi & \sin\phi\end{bmatrix}. \tag{20}$$

The Schrödinger equation in the new basis reads

$$i\frac{d}{dt}\mathbf{C}'(t)=\mathsf{H}'(t)\mathbf{C}'(t), \tag{21}$$

with $\mathbf{C}'(t)=[C_1'(t),C_2'(t),C_3(t)]^T$ and (an overdot meaning a time derivative)

$$\mathsf{H}'=\mathsf{R}^{-1}\mathsf{H}\mathsf{R}-i\mathsf{R}^{-1}\dot{\mathsf{R}}=\begin{bmatrix}\Delta_1' & \Omega'-i\dot\theta\sin\phi & -i\dot\theta\cos\phi\\ \Omega'+i\dot\theta\sin\phi & \Delta_2' & i\dot\phi\\ i\dot\theta\cos\phi & -i\dot\phi & \Delta_3'-\frac{1}{2}i\Gamma\end{bmatrix}, \tag{22}$$

where Γ is given by Eq. (12), and

$$\Delta_1'=\frac{1}{2(\Gamma_1+\Gamma_2)}[\Gamma_3(q_{23}\Gamma_1+q_{13}\Gamma_2)+q_{12}(\Gamma_1+\Gamma_2)^2-(\Gamma_1+\Gamma_2)(q_{13}\Gamma_1+q_{23}\Gamma_2)], \tag{23a}$$

$$\Delta_2'=\frac{\Gamma_3(q_{13}\Gamma_1+q_{23}\Gamma_2)}{2(\Gamma_1+\Gamma_2)}, \tag{23b}$$

$$\Delta_3'=-\frac{1}{2}(q_{13}\Gamma_1+q_{23}\Gamma_2), \tag{23c}$$

$$\Omega'=\frac{q_{13}-q_{23}}{2(\Gamma_1+\Gamma_2)}\sqrt{\Gamma_1\Gamma_2\Gamma_3(\Gamma_1+\Gamma_2+\Gamma_3)}. \tag{23d}$$

Note that $\cot 2\chi=(\Delta_2'-\Delta_1')/2\Omega'$.

III. COINCIDENT PULSES

A. Case of equal Fano parameters

The above theory allows us to derive analytic formulas for the bound-state populations and the ionization probability in the case when all ionization rates have the same time dependence:

$$\Gamma_k(t)=\gamma_k f(t)\quad (k=1,2,3). \tag{24}$$

Then the mixing angles θ, ϕ, and χ are constant, and the nonadiabatic couplings (which are proportional to derivatives of these angles) vanish identically. The solution can be found by an appropriate change of the independent variable (time), and transformation to the adiabatic basis where all nonadiabatic couplings vanish and the Hamiltonian is diagonal. Let us also assume for simplicity that all Fano parameters are equal: $q_{12}=q_{13}=q_{23}\equiv q$. If the population is ini-

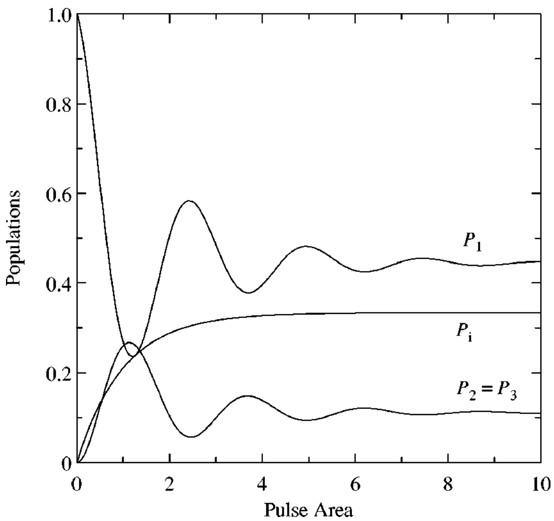

FIG. 2. The populations of the bound states and the ionization probability (25) in the case of coincident pulses [Eq. (24)] plotted against the (dimensionless) pulse area A. All Fano parameters are equal, $q_{12}=q_{13}=q_{23}=5$, and all ionization rates are also equal $(\gamma_1=\gamma_2=\gamma_3)$.

tially in state ψ_1, the populations of the bound states and the ionization after the interaction are easily found to be

$$P_1=\frac{1}{\gamma^2}\left[(\gamma_2+\gamma_3)^2+\gamma_1^2 e^{-A}+2\gamma_1(\gamma_2+\gamma_3)e^{-A/2}\cos\frac{1}{2}qA\right], \tag{25a}$$

$$P_2=\frac{\gamma_1\gamma_3}{\gamma^2}\left(1+e^{-A}-2e^{-A/2}\cos\frac{1}{2}qA\right), \tag{25b}$$

$$P_3=\frac{\gamma_1\gamma_2}{\gamma^2}\left(1+e^{-A}-2e^{-A/2}\cos\frac{1}{2}qA\right), \tag{25c}$$

$$P_i=\frac{\gamma_1}{\gamma}(1-e^{-A}), \tag{25d}$$

where $\gamma=\gamma_1+\gamma_2+\gamma_3$ and

$$A=\int_{-\infty}^{\infty}\Gamma(t)dt. \tag{26}$$

The results are similar when the system is initially in state ψ_2 or ψ_3; then the initial-state population is given by Eq. (25a), the populations of the other two states by Eqs. (25b) and (25c), and the ionization by Eq. (25d) (with an appropriate permutation of the labels). Obviously, a similar population trapping phenomenon as for a two-state LICS takes place, limiting the maximum possible ionization probability to $\frac{1}{3}$ for equal ionization rates $(\gamma_1=\gamma_2=\gamma_3)$, compared to $\frac{1}{2}$ for two-state LICS.

In Fig. 2, populations (25) are plotted against the pulse area A for the case of equal ionization rates. As the pulse area increases, the populations tend to their adiabatic limits $P_1\to\frac{4}{9}$, $P_2=P_3\to\frac{1}{9}$, and $P_i\to\frac{1}{3}$.

B. General case

In the general case of unequal Fano parameters one can still find an analytic solution, but the resulting formulas are too cumbersome to be presented here. The qualitative behavior of the populations remains essentially the same. A simple estimate exists for the maximum possible ionization probability (achieved in the limit of strong ionization rates), which is equal to the initial population of the only decaying adiabatic state $\varphi_3(t)$ [Eq. (16c)]:

$$P_{i,\max}=\sin^2\theta\cos^2\phi=\frac{\gamma_1}{\gamma_1+\gamma_2+\gamma_3}. \tag{27}$$

Hence, the stronger the Stokes and control pulses Γ_2 and Γ_3, the smaller the ionization.

IV. DELAYED PULSES

A. Minimal and maximal ionization

1. No ionization

It is easily seen from Eq. (16c) that when $\theta(-\infty)=0$ and/or $\phi(-\infty)=\frac{1}{2}\pi$, the only decaying adiabatic state $\varphi_3(t)$ is not populated initially. As Eqs. (17a) and (17b) show, this happens when

$$\lim_{t\to-\infty}\frac{\Gamma_1(t)}{\Gamma_1(t)+\Gamma_2(t)+\Gamma_3(t)}=0. \tag{28}$$

In the adiabatic limit state $\varphi_3(t)$ remains unpopulated and hence, the ionization probability is zero throughout the interaction, $P_i(t)=0$. In other words, in the adiabatic limit the ionization probability is zero when the pump pulse is delayed with respect to the Stokes pulse and/or the control pulse. The pulse ordering (28) generalizes the counterintuitive pulse order in the two-state LICS and provides the most appropriate conditions for coherent processes via the continuum, such as population transfer between the bound states, which we shall discuss in Sec. IV B.

2. Complete ionization

As follows from Eq. (16c), when $\theta(-\infty)=\frac{1}{2}\pi$ and $\phi(-\infty)=0$, the decaying state $\varphi_3(t)$ is the only adiabatic state populated initially. According to Eqs. (17a) and (17b), this happens when the pump pulse $\Gamma_1(t)$ arrives before both the control and Stokes pulses, i.e.,

$$\lim_{t\to-\infty}\frac{\Gamma_2(t)}{\Gamma_1(t)}=\lim_{t\to-\infty}\frac{\Gamma_3(t)}{\Gamma_1(t)}=0. \tag{29}$$

In the adiabatic regime no population is transferred to the other adiabatic states, and the ionization probability is given by $P_i=1-|\varphi_3(+\infty)|^2$. Since the decay rate of state $\varphi_3(t)$ is $\frac{1}{2}\Gamma(t)$ [see Eq. (11)], we find that

$$P_i=1-e^{-A}, \tag{30}$$

where A is given by Eq. (26), i.e., P_i can approach unity for strong ionization rates, even though the trapping conditions

(13) are satisfied. The pulse order (29) generalizes the intuitive pulse order in the two-state LICS.

B. Population transfer via continuum

1. Adiabatic limit

An intriguing process based on a LICS is population transfer between two bound states via a common continuum, which has received considerable attention recently [27,28,33–40]. We will show that the tripod system enables the same process, providing at the same time a greater flexibility.

Let us consider the pulse timing when the control pulse $\Gamma_3(t)$ arrives first and disappears last, i.e.,

$$\lim_{t\to\pm\infty}\frac{\Gamma_1(t)}{\Gamma_3(t)}=\lim_{t\to\pm\infty}\frac{\Gamma_2(t)}{\Gamma_3(t)}=0. \tag{31a}$$

As we have shown above (Sec. IV A 1), the ionization probability in this case is zero, $P_i(t)=0$, because the only decaying adiabatic state $\varphi_3(t)$ is not populated initially. Hence the population is distributed among the bound states throughout the interaction. Suppose also that the Stokes pulse precedes the pump pulse (counterintuitive order), i.e.,

$$\lim_{t\to-\infty}\frac{\Gamma_1(t)}{\Gamma_2(t)}=0, \quad \lim_{t\to+\infty}\frac{\Gamma_2(t)}{\Gamma_1(t)}=0. \tag{31b}$$

It follows from Eqs. (17) that

$$\theta(-\infty)=0, \quad \theta(+\infty)=\frac{1}{2}\pi, \tag{32a}$$

$$\phi(-\infty)=\frac{1}{2}\pi, \quad \phi(+\infty)=\frac{1}{2}\pi. \tag{32b}$$

The initial and final values of χ, however, depend on the Fano parameters.

For $q_{13}=q_{23}\neq q_{12}$, we have $\chi(\pm\infty)=0$. Hence

$$\varphi_1(-\infty)=\psi_1, \quad \varphi_1(+\infty)=-\psi_2, \tag{33a}$$

$$\varphi_2(-\infty)=\psi_2, \quad \varphi_2(+\infty)=\psi_1, \tag{33b}$$

$$\varphi_3(-\infty)=\psi_3, \quad \varphi_3(+\infty)=\psi_3. \tag{33c}$$

Thus, in the adiabatic limit, the population is transferred from state ψ_1 to state ψ_2 via the adiabatic state $\varphi_1(t)$.

For $q_{13}\neq q_{23}$, we have $\chi(-\infty)=\frac{1}{2}\pi$ and $\chi(+\infty)=0$. Hence

$$\varphi_1(\pm\infty)=-\psi_2, \tag{34a}$$

$$\varphi_2(\pm\infty)=\psi_1, \tag{34b}$$

$$\varphi_3(\pm\infty)=\psi_3. \tag{34c}$$

Thus, in the adiabatic limit, the population returns to the initial state ψ_1, staying all the time in the adiabatic state $\varphi_2(t)$.

For $q_{13}=q_{23}=q_{12}$, the population behavior is most easily revealed in the $(\varphi_1',\varphi_2',\varphi_3)$ basis because there $\Delta_1'(t)-\Delta_2'(t)=0$ and $\Omega'(t)=0$ [see Eqs. (23)]. Hence states $\varphi_1'(t)$ and $\varphi_2'(t)$ are degenerate, and the coupling between them is given by $\dot{\theta}(t)\sin\phi(t)$. For pulse ordering (31), states $\varphi_1'(t)$ and $\varphi_2'(t)$ have the following asymptotic behaviors [see Eqs. (18)]:

$$\varphi_1'(-\infty)=\psi_1, \quad \varphi_1'(+\infty)=-\psi_2, \tag{35a}$$

$$\varphi_2'(-\infty)=\psi_2, \quad \varphi_2'(+\infty)=\psi_1. \tag{35b}$$

Hence the bare-state populations in the adiabatic limit are

$$P_1\approx\sin^2\int_{-\infty}^{\infty}\dot{\theta}(t)\sin\phi(t)dt, \tag{36a}$$

$$P_2\approx\cos^2\int_{-\infty}^{\infty}\dot{\theta}(t)\sin\phi(t)dt, \tag{36b}$$

$$P_3\approx 0. \tag{36c}$$

The populations of states ψ_1 and ψ_2 depend only on the angles $\theta(t)$ and $\phi(t)$, which in turn depend on the time delay τ between $\Gamma_1(t)$ and $\Gamma_2(t)$. This dependence provides the possibility to control the created coherent superposition of ψ_1 and ψ_2 through the pulse delay. This property of the tripod-continuum system is similar to the one for a discrete tripod system coupled via a discrete state [41], rather than a continuum, which was demonstrated experimentally recently [42].

2. Optimal conditions for population transfer

Although in the general case of $q_{13}\neq q_{23}$ the population returns to the initial state ψ_1 in the adiabatic limit, it is still possible to transfer population to state ψ_2 for certain ranges of interaction parameters. These ranges are most easily determined in the $(\varphi_1',\varphi_2',\varphi_3)$ basis, which is more convenient than the adiabatic basis. As is evident from the asymptotic limits [Eqs. (35)] of $\varphi_1'(t)$ and $\varphi_2'(t)$, only state $\varphi_1'(t)$ is populated initially, and if the atom stays in $\varphi_1'(t)$ at all times, the desired population transfer from ψ_1 to ψ_2 will occur. In order to achieve this, transitions from $\varphi_1'(t)$ to both states $\varphi_2'(t)$ and $\varphi_3(t)$ must be suppressed. This restriction determines the ranges of interaction parameters for which significant population transfer from ψ_1 to ψ_2 is possible.

State $\varphi_1'(t)$ is coupled to state $\varphi_3(t)$ with a coupling proportional to $\dot{\theta}(t)$ [see Eqs. (22)]. Hence the detrimental transitions from $\varphi_1'(t)$ to $\varphi_3(t)$ can be avoided if the interaction is sufficiently adiabatic, which requires that

$$|\dot{\theta}(t)\cos\phi(t)|\ll\sqrt{[\Delta_1'(t)-\Delta_3'(t)]^2+\frac{1}{4}\Gamma^2(t)}. \tag{37}$$

On the other hand, the interaction should not be too adiabatic, because then, as we have shown in Sec. IV B 1, the population returns to state ψ_1. This conclusion is confirmed when examining the nature of the interaction between states $\varphi_1'(t)$ and $\varphi_2'(t)$ [see Eq. (22)]. Indeed, the effective detuning in this subsystem $\Delta_2'(t)-\Delta_1'(t)$ has different signs at $t\rightarrow\pm\infty$, which means that there is a level-crossing transition and hence, complete population transfer between states $\varphi_1'(t)$ and $\varphi_2'(t)$ occurs in the adiabatic limit. According to Eqs. (35), such a complete transfer means complete population return to ψ_1 in the bare-state basis. Obviously, only in the case of $q_{13}=q_{23}$ does the coupling $\Omega'(t)$ vanish identically, and $\varphi_1'(t)$ and $\varphi_2'(t)$ are only coupled by a weak nonadiabatic coupling, which vanishes in the adiabatic limit. However, the case $q_{13}=q_{23}$ is exceptional, and it is difficult to find atomic states which satisfy this condition. For $q_{13}\neq q_{23}$, there is a residual coupling $\Omega'(t)$ between $\varphi_1'(t)$ and $\varphi_2'(t)$ which remains nonzero in the adiabatic limit and causes transitions between these states. Referring to the Landau-Zener formula [47], we conclude that in order to avoid population transfer from $\varphi_1'(t)$ to $\varphi_2'(t)$, the relation

$$[\Omega'(t_0)]^2\ll|\dot{\Delta}_2'(t_0)-\dot{\Delta}_1'(t_0)| \tag{38}$$

must be fulfilled, where t_0 is the crossing point: $\Delta_1'(t_0)=\Delta_2'(t_0)$. It is possible to refine condition (38) by including effects of asymmetry [48] and nonlinearity [49] at the crossing and finite transition times [50].

Conditions (37) and (38) provide the restrictions on the interaction parameters needed for significant population transfer from ψ_1 to ψ_2.

3. Numerical examples

In our numerical simulations we have used Gaussian pulse shapes for $\Gamma_1(t)$ and $\Gamma_2(t)$ and constant Γ_3,

$$\Gamma_1(t)=\gamma_1 e^{-(t-\tau)^2/T^2}, \tag{39a}$$

$$\Gamma_2(t)=\gamma_2 e^{-(t+\tau)^2/T^2}, \tag{39b}$$

$$\Gamma_3(t)=\gamma_3, \tag{39c}$$

where 2τ is the delay between the pump and Stokes pulses and T is their width.

It is possible to simplify conditions (37) and (38) when the control pulse is much stronger than the pump and Stokes pulses, and the latter two have equal peak values: $\gamma_3\gg\gamma_1=\gamma_2$. Then the crossing point is given by $t_0\approx 0$, and conditions (37) and (38) become

$$\frac{2\tau}{T\sqrt{1+\frac{1}{4}(q_{13}+q_{23})^2}}\ll\gamma_3 T\ll\frac{16\tau}{T|q_{13}-q_{23}|}. \tag{40}$$

Hence an appreciable population transfer from ψ_1 to ψ_2 is only possible if the difference $|q_{13}-q_{23}|$ is sufficiently small.

In Fig. 3, the populations of the discrete states and the ionization probability are plotted against the pulse width T of the pump and Stokes pulses. The detunings $\Delta_1(t)$ and $\Delta_2(t)$ are chosen to satisfy the trapping conditions (13) at any time;

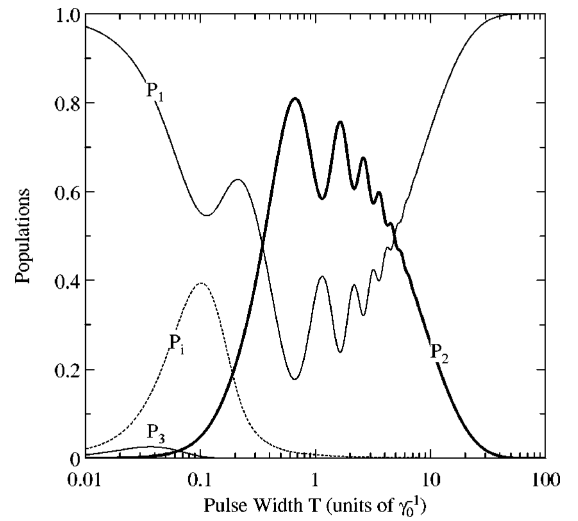

FIG. 3. The populations of the discrete states and the ionization probability plotted against the pulse width T of the pump and Stokes pulses. The pulse shapes are given by Eqs. (39) with $\tau=0.5T$, $\gamma_1=\gamma_2\equiv\gamma_0$, and $\gamma_3=3\gamma_0$. We have chosen the maximum ionization rate γ_0 for states ψ_1 and ψ_2 to determine the frequency and time scales. The Fano parameters are $q_{13}=5$, $q_{23}=5.5$, and $q_{12}=2$. The detunings $\Delta_1(t)$ and $\Delta_2(t)$ are assumed to satisfy the trapping conditions (13) at any time.

as noted in Sec. I, this can be achieved, at least in principle, by using the Stark shifts induced by an additional (nonionizing) laser [36] or by using appropriately chirped laser pulses [37,38]. In this case, the Stark shifts $S_k(t)(k=1,2,3)$ are unimportant because they enter Eq. (1) through $\Delta_1(t)$ and $\Delta_2(t)$ only [which are given the values prescribed by Eqs. (13)], and are therefore set equal to zero. The figure shows that a reasonably high efficiency of population transfer to state ψ_2 can be achieved in a certain range of T; this range is predicted correctly by condition (40), which in this case reads as $0.2\ll\gamma_3 T\ll 16$. For small T, the interaction is nonadiabatic and the population is distributed mainly between the initial state (due to a transition from φ_1' to φ_2') and the continuum (due to a transition from φ_1' to φ_3). As T increases, the interaction becomes increasingly adiabatic and the ionization probability P_i is reduced, as well as the initial-state population P_1. For large T the interaction becomes almost completely adiabatic, and the population returns to the initial state because of the level-crossing transition from φ_1' to φ_2'.

As Eqs. (13) show, for large and constant Γ_3, the trapping conditions are satisfied approximately at $\delta_1\approx\frac{1}{2}q_{13}\Gamma_3$ and $\delta_2\approx\frac{1}{2}q_{23}\Gamma_3$. The implication is that in this case it may be easier to satisfy the two (constant) trapping conditions for the tripod system than the single (time-dependent) trapping condition (14) for the two-state LICS. In Fig. 4, the population of state ψ_2 is plotted against the sum and the difference of the detunings δ_1 and δ_2 for three values of the constant ionization rate γ_3. For $\gamma_3=0$, when state ψ_3 is uncoupled, P_2 depends only on the two-photon detuning $\delta_1-\delta_2$ between states ψ_1 and ψ_2, as expected. The figure shows that for $\gamma_3=\gamma_0$ and $\gamma_3=4\gamma_0$, there is a region in the (δ_1,δ_2) plane, where P_2 achieves higher values than for $\gamma_3=0$ (and

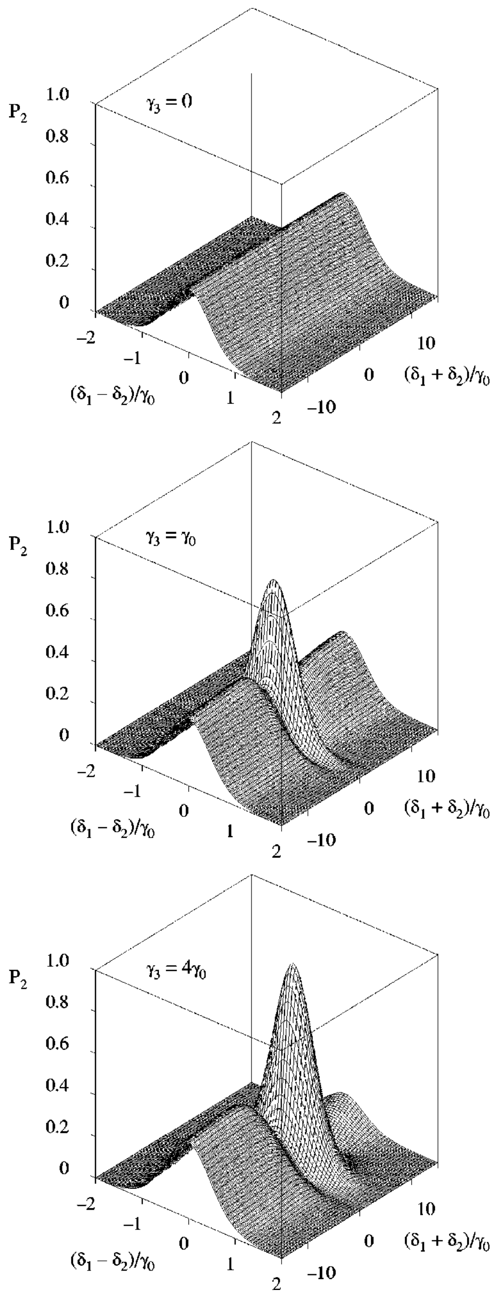

FIG. 4. The population of state ψ_2 plotted against the sum and the difference of the detunings δ_1 and δ_2 (in units γ_0) for three different constant ionization rates: $\gamma_3=0$ (upper frame), $\gamma_3=\gamma_0$ (middle frame), and $\gamma_3=4\gamma_0$ (lower frame). The pulse shapes are given by Eqs. (39) with $\tau=0.5T$, $\gamma_1=\gamma_2\equiv\gamma_0$, and $\gamma_0 T=1$. The Fano parameters are $q_{13}=1$, $q_{23}=1.2$, and $q_{12}=2$. The Stark shifts of all states are neglected. As in Fig. 3, we have chosen the maximum ionization rate γ_0 for states ψ_1 and ψ_2 to determine the frequency and time scales.

also regions where P_2 achieves lower values). The maximum transfer efficiency is approximately 0.30 for $\gamma_3=0$, 0.76 for $\gamma_3=\gamma_0$, and 0.95 for $\gamma_3=4\gamma_0$. This shows that, indeed, in a certain detuning range it is easier to satisfy the two trapping conditions (13) for the tripod system than the single trapping condition for the two-state LICS. In this example, the Stark shifts $S_k(t)$ $(k=1,2,3)$ of the three bound states were neglected and set to zero. Their inclusion would not introduce qualitative changes [because the trapping conditions (13) are not satisfied anyway, even at the maxima], but could only slightly modify the values of P_2.

V. EFFECTIVE TWO-STATE LICS SYSTEM

Finally, we discuss the case when Γ_3 is large compared to Γ_1 and Γ_2. For example, such a situation arises when state ψ_3 is an autoionizing state whose coupling to the continuum (by configuration interaction) is usually much stronger than laser ionization rates. Then we can eliminate state ψ_3 adiabatically by setting $dC_3/dt=0$ in Eq. (1), determining C_3 in terms of C_1 and C_2 from the resulting algebraic equation, and replacing C_3 in the other two equations. We also make a (population preserving) phase transformation that shifts the zero energy level to coincide with the modified energy of state ψ_1. We thus reduce the initial three-state problem to an effective two-state one, involving states ψ_1 and ψ_2 only,

$$i\frac{d}{dt}\mathbf{C}^{\text{ae}}(t)=\mathsf{H}^{\text{ae}}(t)\mathbf{C}^{\text{ae}}(t), \tag{41}$$

where $\mathbf{C}^{\text{ae}}(t)=[C_1^{\text{ae}}(t),C_2^{\text{ae}}(t)]^T$ and

$$\mathsf{H}^{\text{ae}}(t)=\frac{1}{2}\begin{bmatrix} -i\Gamma_1^{\text{ae}} & -\sqrt{\Gamma_1^{\text{ae}}\Gamma_2^{\text{ae}}}(q^{\text{ae}}+i) \\ -\sqrt{\Gamma_1^{\text{ae}}\Gamma_2^{\text{ae}}}(q^{\text{ae}}+i) & 2\Delta^{\text{ae}}-i\Gamma_2^{\text{ae}} \end{bmatrix}, \tag{42}$$

with

$$\Gamma_1^{\text{ae}}(t)=\Gamma_1(t)q_{13}^2, \tag{43a}$$

$$\Gamma_2^{\text{ae}}(t)=\Gamma_2(t)q_{23}^2, \tag{43b}$$

$$q^{\text{ae}}=\frac{q_{12}-q_{13}-q_{23}}{q_{13}q_{23}}, \tag{43c}$$

$$\Delta^{\text{ae}}(t)=\delta_2-\delta_1+\Sigma_2(t)-\Sigma_1(t)+\Gamma_2(t)q_{23}-\Gamma_1(t)q_{13}, \tag{43d}$$

where the label ''ae'' stands for ''adiabatic elimination.'' Hence we obtain a standard two-state LICS problem with modified ionization rates $\Gamma_1^{\text{ae}}(t)$ and $\Gamma_2^{\text{ae}}(t)$, Fano parameter q^{ae}, and detuning $\Delta^{\text{ae}}(t)$. Thus the presence of a third state, strongly coupled to the continuum, modifies the properties of the two-state problem involving states ψ_1 and ψ_2. It may happen that the modified parameters, and in particular q^{ae}, have more suitable values for observing and investigating LICS and related phenomena, such as population transfer. In particular, if the Fano parameters q_{13}, q_{23}, and q_{12} are large, the effective Fano parameter q^{ae} will be small, which can facilitate the observation of a LICS [17,18].

It is possible to obtain further insight of the tripod-continuum system by adiabatic elimination of the only decaying adiabatic state $\varphi_3(t)$ both in the adiabatic basis and in the $(\varphi_1',\varphi_2',\varphi_3)$ basis.

VI. SUMMARY AND CONCLUSIONS

In the present paper we have investigated the coherence properties of a system involving three discrete states coupled to each other by two-photon processes via a common continuum. In this tripod scheme, there exist two population trapping conditions, rather than just one as in a standard LICS. In some cases, e.g., for a strong and constant control pulse, it may be easier to satisfy these conditions than the single trapping condition in a standard LICS. Depending on the pulse timing, various effects can be observed. We have derived some basic properties of the tripod scheme, such as the solution for coincident pulses (sharing the same time dependence), the behavior of the system in the adiabatic limit for delayed pulses, the conditions for no ionization and for maximal ionization, and the optimal conditions for population transfer between the discrete states via the continuum. In the case of a strongly coupled state, by adiabatically eliminating this state, we have found that the tripod scheme reduces to an effective standard two-state LICS system with modified Fano parameter and ionization rates; such modification may provide better conditions for observing and investigating a LICS and related phenomena.

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