Adiabatic passage and dissociation controlled by interference of
two laser-induced continuum structures

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Abstract

We have developed a theory of three-pulse coherent control of photochemical processes. It is based on adiabatic passage and quantum coherence and interference attributed to the lower-lying dissociation continuum and excited upper discrete states, which are otherwise not connected to the ground state by one-photon transitions. Novel opportunities offered by the proposed scheme are demonstrated through extensive numerical simulations with the aid of a model relevant to typical experiments. The opportunities for manipulating the distribution of the population among discrete and continuous states with any necessary ratio by the end of the pulses are demonstrated.

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I. INTRODUCTION

Laser control of chemical reactions and other dissipation processes like photodissociation of molecules and photoionization of atoms has recently gained increasing interest in the laser and chemical communities [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19]. In many cases, such control is based on the interference of different quantum pathways. While the main stream of theoretical and experimental work on coherent quantum control concerns laser-induced transitions between discrete states, coherence and interference effects attributed to dissociation and ionization energy continua are of great importance, because of various applications in atomic and molecular physics and photochemistry. The properties of the continuum of quantum states such as observed in the ionization of an atom or dissociation of a molecule have attracted interest since the first formulation of quantum theory. Despite the fact that the shape of an autoionizing resonance is stipulated by quantum interference [20, 21], for a long time the continuum was regarded as an incoherent dissipation medium. It was until the work [22, 23, 24, 25, 26] which established that the mixing of discrete and continuous states in a strong, driving electromagnetic field led to similar constructive and destructive interference processes. Thus the opportunity was shown to produce an autoionizing-like resonance embedded in an otherwise unstructured continuum, which displays itself in the variety of photo- and nonlinear-optical processes similar to real autoionizing states [24, 25, 26, 27, 28, 29]. The attractive advantage of this option is that such laser-induced continuum structures (LICS) can be produced, at least in principal, in any necessary area of a continuum, and their strength is controlled with the dressing laser. Since the first successful experiments [30, 31, 32, 33], which confirmed the principal theoretical predictions, a great progress has been achieved in developing a profound understanding of continuum coherence and related laser-induced processes [34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49, 50, 51, 52, 53, 54, 55, 56, 57, 58, 59].

The appearance and consequences of coherence and interference processes are different in the continuous wave regime, where the relaxation processes play a crucial role, and in the pulsed regime of rapid adiabatic passage [60, 61, 62, 63, 64, 65, 66, 67, 68, 69, 70, 71, 72, 73, 74, 75, 76, 77, 78, 79]. Extensive studies of population transfer between two discrete states via the upper-lying continuum in Raman-like Λ-schemes have revealed that, despite the detrimental effects, almost complete population transfer and corresponding photoionization
suppression is possible in such schemes, provided that the driving laser pulses are properly ordered [80, 81, 82, 83, 84, 85, 86, 87, 88]. It was also found that coherence processes play an important role in two-photon dissociation mediated by an intermediate resonance with a discrete level allowing for the control of photodissociation and photoassociation [89, 90, 91, 92, 93, 94]. Population transfer and dissociation play an important role as the accompanying processes in the three-pulse four-wave mixing technique, which is used for spectroscopy and coherent quantum control in chemistry [95, 96, 97, 98, 99, 100, 101].

Based on the outlined achievements, this paper further develops the theory of coherent quantum control, which employs LICS and rapid adiabatic passage. We propose and investigate a more complex three-pulse scheme (Fig. 1), which allows additional flexibility and means for such control. Unlike the so far investigated schemes, we report on novel opportunities for manipulating dissociation and population redistributions between the upper bound states \( n \) and \( f \) through the interplay of two LICS attributed to lower-lying energy continuum. Only one discrete state here is connected to the ground state by one-photon transitions. All radiations are assumed to be Gaussian pulses with variable sequence and duration, strong enough to drive molecular transitions. Corresponding equations for slowly-varying probability amplitude are derived, and extensive numerical simulations are performed aimed at a demonstration of the outlined opportunities under typical experimental conditions.

This paper is organized as follows. The set of equations for slowly-varying probability amplitudes pertinent to the problem under consideration are derived in Sec. II. By turning off the first or third laser, our scheme can be easily reduced to those investigated in [85, 94], where excellent agreement between theory and experiment was reported. Corresponding analytical solutions are found for further analysis of time-dependent destructive and constructive interference and for testing the theory by comparing with the known results. They are also useful for determining the dependence of such processes on the complex effective Fano parameters, which represent the properties of the specific continua. The results of various numerical experiments are presented in in Sec. III. Suppression of photodissociation through continuum coherence and overlap of two LICS is simulated in Sec. III A. The features of photodissociation controlled by two ordered pulses are simulated in Sec. III B. We investigate the possibility of manipulating the dissociation yield and populations of excited states, driven by three pulses with variable sequence and duration. Our goal is to demonstrate the feasibility of achieving almost any necessary ratio between the branching yields.
by the end of the pulses due to the interference of quantum pathways through a variety of continuum states. This is done with the aid of numerical experiments in Sec. III C. We show how the pulse parameters must be adjusted in order to enable such opportunities.

II. BASIC EQUATIONS

The proposed coupling scheme is illustrated in the energy level diagram depicted in Fig. 1. Radiation at frequency $\omega_1$ couples the bound-bound transition $m - n$, and radiations at $\omega_2$ and $\omega_3$ couple the bound states $n$ and $f$ with states of the dissociation continuum $\varepsilon$, as shown in the picture. Initially, only the lowest level is assumed populated. At $\omega_1$ tuned near the one-photon resonance, stepwise and two-photon transitions interfere. The two-photon transition rate between the levels $n$ and $f$ depends on the detuning $\Omega_{nf}$. Actually, all detunings are intensity-dependent, which determine the complex dynamics of the interference between various stepwise and multiphoton processes. We assume that each laser drives only one transition and accounts only for resonant coupling depicted in Fig. 1. Therefore, we ignore other off-resonant and incoherent processes caused by the driving field. We also assume a single continuum, i.e., no degeneracy, in order to avoid unnecessary complexity related to the processes, which are not the subject of this paper. Basically, the set of equations depends on whether the system is open or closed. In the case of a closed model, the lowest level $m$ is the ground state, and the sum of the level populations is always equal to 1. Alternatively, in an
open scheme, level $m$ is an excited state that relaxes, and the sum of the populations depletes with time. For the purposes of discussion, we will consider the second case, although for pulses considerably shorter than all the relaxation times, the difference between the closed and open schemes disappears. In the rotating-wave approximation, the equations for slowly-varying probability amplitudes in the case of an open energy-level system are written as

$$
i a_\varepsilon = G_{\varepsilon n} \exp(i\Omega_{\varepsilon n} t)a_n + G_{\varepsilon f} \exp(i\Omega_{\varepsilon f} t)a_f$$
$$i \dot{a}_n + i\gamma_n a_n = G_{nm} \exp(i\Omega_{nm} t)a_m + \int d\varepsilon G_{n\varepsilon} \exp(i\Omega_{n\varepsilon} t)a_\varepsilon,$$
$$i \dot{a}_f + i\gamma_f a_f = \int d\varepsilon G_{f\varepsilon} \exp(i\Omega_{f\varepsilon} t)a_\varepsilon,$$
$$i \dot{a}_m + i\gamma_m a_m = G_{mn} \exp(i\Omega_{mn} t)a_n.$$  

(1)

Here $G_{\varepsilon n} = E_2 d_{\varepsilon n}/2\hbar$, $G_{\varepsilon f} = E_3 d_{\varepsilon f}/2\hbar$, and $G_{mn} = E_1 d_{mn}/2\hbar$ are coupling Rabi frequencies; $\Omega_{mn} = \omega_1 - \omega_{mn}$, $\Omega_{n\varepsilon} = \omega_2 - \omega_{n\varepsilon}$ and $\Omega_{f\varepsilon} = \omega_3 - \omega_{f\varepsilon}$ are frequency resonance detunings; $\omega_{i\varepsilon} = (E_i - \varepsilon)/\hbar$; $E_i$ and $\varepsilon$ are the energies of the corresponding discrete and continuum states; and $2\gamma_i$ is the decay rate of level $i$. Following [29], we introduce the values

$$a_\varepsilon(t) = \bar{a}_\varepsilon \exp(i\Omega_{\varepsilon \varepsilon_0} t), \quad a_n(t) = \bar{a}_n \exp(i\Omega_{n\varepsilon_0} t),$$
$$a_f(t) = \bar{a}_f \exp(i\Omega_{f\varepsilon_0} t), \quad a_m(t) = \bar{a}_m \exp(i\Omega_{m\varepsilon_0} t),$$

(2)

where $\bar{a}_\varepsilon$ is the amplitude, which slowly varies over the continuum in the vicinity of the energy $\varepsilon_0 = E_f - \hbar\omega_3$,

$$\Omega_{f\varepsilon_0} = \omega_{f\varepsilon_0} - \omega_3 = 0, \quad \Omega_{n\varepsilon_0} = \omega_{n\varepsilon_0} - \omega_2 = \omega_{f\varepsilon_0} - \omega_n - \omega_2 = \Omega_{nf},$$
$$\Omega_{m\varepsilon_0} = \omega_{m\varepsilon_0} - \omega_1 + \omega_2 = \omega_{mn} - \omega_{m\varepsilon_0} - \omega_1 + \omega_2 = \Omega_{mn} - \Omega_{n\varepsilon}.$$  

(3)

The energy $\varepsilon_0$ is chosen in the vicinity of $E_n - \hbar\omega_2$, if $E_3 = 0$. We assume that the coupling parameters (energy density of the transition oscillator strengths $f_{i\varepsilon}$) vary around $\varepsilon_0$ substantially only over the continuum energy intervals $\bar{\varepsilon}$, which are much greater than the maximum of the characteristic widths of the discrete states, including power-broadening $\hbar|V_{jk}|$ and pulse spectral width $\tau^{-1}$:

$$\bar{\varepsilon} \gg \hbar\gamma_i, \hbar/\tau, \quad \hbar|V_{jk}| \partial^2 f/\partial \varepsilon^2_{\varepsilon = \varepsilon_0} \ll \partial f/\partial \varepsilon_{\varepsilon = \varepsilon_0}.$$  

(4)

Such requirements are fulfilled practically for all realistic atomic and molecular continua in an energy range that is well above the ionization or dissociation threshold. Since interference
processes appear at \(|\Omega_{mn}|, |\Omega_{nf}|\) and \(|\Omega_{mn} - \Omega_{nf}|\) less than or on the order of the same value, this enables us to separate the slow and fast variables. With the aid of (2), we obtain from (1)

\[
\dot{a}_\varepsilon - \Omega_{\varepsilon\varepsilon_0} a_\varepsilon = G_{\varepsilon n} a_n + G_{\varepsilon f} a_f, \tag{5}
\]

\[
\dot{\pi}_n - \pi_n (\Omega_{nf} - i\gamma_n) = G_{nm} \pi_m + \int d\varepsilon G_{n\varepsilon} \pi_\varepsilon, \tag{6}
\]

\[
\dot{a}_f + i a_f \gamma_f = \int d\varepsilon G_{f\varepsilon} a_\varepsilon, \tag{7}
\]

\[
\dot{\pi}_m - \pi_m (\Omega_{mn} - \Omega_{nf} - i\gamma_m) = G_{mn} \pi_n. \tag{8}
\]

The solution of (5) is

\[
a_\varepsilon = e^{-i\Omega_{\varepsilon\varepsilon_0} t} [C_1 + \int (G_{\varepsilon n} a_n + G_{\varepsilon f} a_f) e^{i\Omega_{\varepsilon\varepsilon_0} t} dt].
\]

Taking slowly-varying values \(G_{\varepsilon i} a_i\) (as compared with the oscillating exponents) out of the integral, we obtain

\[
a_\varepsilon = e^{-i\Omega_{\varepsilon\varepsilon_0} t} [C_1 + (G_{\varepsilon n} a_n + G_{\varepsilon f} a_f) \int e^{i\Omega_{\varepsilon\varepsilon_0} t} dt] = e^{-i\Omega_{\varepsilon\varepsilon_0} t} C_1 - (G_{\varepsilon n} a_n + G_{\varepsilon f} a_f)/\Omega_{\varepsilon\varepsilon_0}.
\]

It follows from the initial conditions that \(C_1 = 0\), and therefore the solution of (5) is

\[
a_\varepsilon = -(G_{\varepsilon n} a_n + G_{\varepsilon f} a_f)/\Omega_{\varepsilon\varepsilon_0}, \tag{9}
\]

This can be derived also directly from (5) because the inequality \(|\dot{a}_\varepsilon| \ll |\Omega_{\varepsilon\varepsilon_0} a_\varepsilon|\) is correct for the major continuum interval. Further, with the aid of the \(\zeta\)-function,

\[
[i(\omega_k - \omega_{je})]^{-1} = \pi \delta(\omega_k - \omega_{je}) - i \mathcal{P} (\omega_k - \omega_{je})^{-1}; \quad k = 1, 2, 3; \quad j = m, n, f, \tag{10}
\]

where \(\mathcal{P}\) stands for the principal value of an integral, Eqs. (6)-(8) can be presented in the form

\[
\frac{d\pi_m}{dt} = -iG_{mn} \pi_n - (\gamma_m + i\Omega_{m\varepsilon_0})\pi_m,
\]

\[
\frac{d\pi_n}{dt} = -iG_{nm}^* \pi_m - \gamma_{nf}(1 + iq_{nf}) \pi_f - [\gamma_n + \gamma_{mn} + i(\Omega_{n\varepsilon_0} + \delta_{nn})] \pi_n, \tag{11}
\]

\[
\frac{d\pi_f}{dt} = -\gamma_{fn}(1 + iq_{fn}) \pi_n - (\gamma_f + \gamma_{ff} + i\delta_{ff}) \pi_f.
\]
where

\[
\gamma_{nn} = \pi \hbar G_{n\varepsilon_0} G_{\varepsilon n} + \text{Re} \left( G_{nk} G_{kn} / p_{kf} \right),
\]

\[
\delta_{nn} = \hbar \mathcal{P} \int d\varepsilon \cdot G_{n\varepsilon} G_{\varepsilon n} / (\varepsilon_0 - \varepsilon) + \text{Im} \left( G_{nk} G_{kn} / p_{kf} \right);
\]

\[
\gamma_{ff} = \pi \hbar G_{f\varepsilon_0} G_{\varepsilon f} + \text{Re} \left( G_{fk} G_{kf} / p_{kf} \right),
\]

\[
\delta_{ff} = \hbar \mathcal{P} \int d\varepsilon \cdot G_{f\varepsilon} G_{\varepsilon f} / (\varepsilon_0 - \varepsilon) + \text{Im} \left( G_{fk} G_{kf} / p_{kf} \right);
\]

\[
\gamma_{nf} = \pi \hbar G_{n\varepsilon_0} G_{\varepsilon f} + \text{Re} \left( G_{nk} G_{kf} / p_{kf} \right),
\]

\[
\delta_{nf} = \hbar \mathcal{P} \int d\varepsilon \cdot G_{n\varepsilon} G_{\varepsilon f} / (\varepsilon_0 - \varepsilon) + \text{Im} \left( G_{nk} G_{kf} / p_{kf} \right);
\]

\[
\gamma_{fn} = \pi \hbar G_{f\varepsilon_0} G_{\varepsilon n} + \text{Re} \left( G_{fk} G_{kn} / p_{kf} \right),
\]

\[
\delta_{fn} = \hbar \mathcal{P} \int d\varepsilon \cdot G_{f\varepsilon} G_{\varepsilon n} / (\varepsilon_0 - \varepsilon) + \text{Im} \left( G_{fk} G_{kn} / p_{kf} \right);
\]

\[
p_{kf} = \Gamma_{kf} + i(\omega_{kf} - \omega_3), \quad q_{ij} = \delta_{ij} / \gamma_{ij}, \quad i, j = n, f.
\]

Besides the continuum states, the contribution of other non-resonant levels \(k\) is taken into account, and a sum over the repeating \(k\) index is assumed. Contributions from these levels may occur comparable to those of the continuum states. As seen from the equations (11), the values \(\gamma_{ij}\) and \(\delta_{ij}\) describe light-induced broadening and shifts of discrete resonances stipulated by the induced transitions between them through the continuum. The magnitude of the shift and its sign are determined by the overall counterbalance of the continuum states below and above \(\varepsilon_0\). The parameters \(q_{ij} = \delta_{ij} / \gamma_{ij}\) are analogous to the Fano parameters for autoionizing states. They characterize the relative integrated contribution of all off-resonant quantum states compared to the resonant ones. Within the validity of (11), their dependence on the field intensities can be neglected. The ratio of the real and imaginary parts (relative phase of the corresponding induced atomic oscillations) plays a crucial role in whether the interference is constructive or destructive. This depends on the effective Fano parameters \(q_{ij}\), which are determined by the distribution of the oscillator strengths over the continuum and discrete states and on the positions of the resonant continuum states, and can be controlled through multiphoton detunings.

Further, we will use dimensionless variables, scaled to the pulse \(|E_1|^2\) half-duration \(\tau\) at the \(1/e\) level: \(T = t / \tau\), \(\gamma_{i\tau} = \gamma_i\), \(g_{mn} = G_{mn\tau}\), \(g_{nf} = \gamma_{nf\tau}\), \(g_{nn} = \gamma_{mn\tau}\), \(g_{ff} = \gamma_{ff\tau}\),
\[ \Delta_{mn} = \Omega_{mn} \tau, \quad \Delta_{nf} = \Omega_{nf} \tau. \]  

Then the equations take the form

\[
\begin{align*}
\frac{d\alpha_m}{dT} &= -ig_{mn}\alpha_n - [\eta_n + i(\Delta_{mn} - \Delta_{nf})]\alpha_m, \\
\frac{d\alpha_n}{dT} &= -ig^*_{mn}\alpha_m - g_{nf}(1 + iq_{nf})\alpha_f - [\eta_n + g_{nn} + i(\Delta_{nf} + q_{nn}g_{nn})]\alpha_n, \\
\frac{d\alpha_f}{d\tau'} &= -g_{fn}(1 + iq_{fn})\alpha_n - (\eta_f + g_{ff} + iq_{ff}g_{ff})\alpha_f.
\end{align*}
\]  

(14)

With the aid of these equations, the dissociation probability is calculated as

\[ W = 2 \int dT \left\{ (g_{mn}\alpha_n^2 + g_{ff}\alpha_f^2 + 2 \text{Re} [g_{nf}\alpha_n\alpha_f^* \exp(i\Delta_{nf}T)]) \right\}. \]  

(15)

The first two terms, proportional to the squared moduli of the probability amplitudes, describe the multistep dissociation associated with the populations of levels \( n \) and \( f \). The third term describes quantum control through the interference of coherent quantum pathways. As seen from Eqs. (11) and (14), the magnitude and sign of the interference term in (15) strongly depends on the phases of the probability amplitudes and, consequently, on the parameter \( q_{fn} \).

### III. NUMERICAL SIMULATION OF TWO- AND THREE-PULSE COHERENT CONTROL

Many experiments on the coherent control of branching chemical reactions have been carried out with sodium dimers Na\(_2\). In this case, level \( m \) of our model (Fig. 1) can be attributed to the state \( X^1\Sigma_g^+(v = 28, J = 10) \), and levels \( n \) and \( f \) to the states \( A^1\Sigma_u^+(v = 37, J = 11) \) and \( A^1\Sigma_u^+(v = 47, J = 11) \), which are coupled by strong transition dipoles with the dissociation continuum Na(3s)+Na(3s) [5]. Alternatively, vibrational states of the electronic excitation \( B^1\Pi_u(v, J = 11) \) can be chosen as level \( f \). The characteristic relaxation rates of these states are: \( \gamma_m = 2 \cdot 10^7 \text{ c}^{-1}, \quad \gamma_n = \gamma_f = 1.2 \cdot 10^8 \text{ c}^{-1}. \) We assume all pulses to be Gaussian,

\[
\begin{align*}
g_{mn} &= g_{mn}^0 \exp(-T^2/2), \\
\gamma_{mn} &= \gamma_{mn}^0 \exp \left[-(T - \Delta_2)^2/d_2^2\right], \\
\gamma_{ff} &= \gamma_{ff}^0 \exp \left[-(T - \Delta_3)^2/d_3^2\right],
\end{align*}
\]  

(16)

where \( \Delta_2 \) and \( \Delta_3 \) are the delays of the pulses \( E_2 \) and \( E_3 \) with respect to the pulse \( E_1 \), \( d_2 = \tau_2/\tau \) and \( d_3 = \tau_3/\tau \) are the durations of pulses \( E_2 \) and \( E_3 \) (\( \tau_2 \) and \( \tau_3 \)) scaled to the
duration of the first pulse $\tau$. These values must be selected within a time domain range shorter than $\tau = 10^{-9}$ s in order to avoid relaxation over the whole period of excitation. As seen from Eq. (13), the overall analysis of the control reduces to an analysis of the interference and multistep terms.

![Graph](image)

FIG. 2: Dissociation (a) and population transfer from level $n$ to level $f$ (b) vs time and delay of the pulse $E_2$ relative to $E_3$ ($\Delta_2 = 0$, $\Delta_3$ is negative ). The field $E_1$ is turned off, the initial population of level $n$ equals 1, $g_{nn}^0 = 3.61$, $g_{ff}^0 = 9.61$, $g_{nf}^0 = 5.89$, the pulse durations are equal and much shorter than the relaxation rates, $\Omega_{nf} = 0$, and the Fano parameters are $q_{nn} = 0.2$, $q_{ff} = -0.5$, $q_{nf} = 10$.

A. Suppression of photodissociation by continuum coherence and overlap of two LICS

Figures 2 and 3 demonstrate feasibilities of manipulating constructive and destructive interference aimed at control of the population transfer between the upper levels $n$ and $f$. Here the first field is turned off and only level $n$ is initially populated. Figure 2 simulates the population dynamics when the carrier frequencies of the fields are tuned to the two-photon resonance with $\omega_{fn}$. It shows a dramatic dependence of the output on the temporal shift between the pulse peaks. Such dynamics is stipulated by the time-dependent shift of the two-photon resonance, which determines constructive or, alternatively, destructive interference. In other words, the position, strength and overlap the two LICS vary in time, which bring about the dynamics and consequences of the interference. For the given parameters, we have almost complete suppression of dissociation [plot (a)] and about 80% population transfer to level $f$ [plot (b)]. This occurs for counterintuitive order of pulses, when third
field reaches its maximum at about four pulse half-duration before the maximum of second pulse. Alternatively, almost complete photodissociation can be ensured with different pulse shifts. Thus our simulations demonstrate a behavior, which is in a good agreement with the conclusions and observations of other studies [85, 86]. Figure 3 [plots (a) and (c)] displays a strong dependence of dissociation and population transfer on the effective Fano parameter $q_{nf}$. This because they determine the magnitude and sign of the resonance shift (position and shape of LICS). The plots (b) and (d) show that similar manipulations can be performed by an appropriate adjustment of the two-photon detuning.

B. Photodissociation controlled by two ordered pulses

Figure 4 shows the features of dissociation from the lowest state controlled with two pulses. Here the third field is turned off, only level $m$ is initially populated, the carrier frequency of the first field is tuned to resonance with the transition $m\nu$, and dissociation is
FIG. 4: Dependence of the two-photon dissociation ($E_3 = 0$) (a,b,c) and population of the ground level (d) on the intensity and pulse delay of the driving fields. $\Omega_{mn} = 0$, $g_{0mn}^0 = 2$. (a) $g_{0nn}^0 = 3.61$; (b) $g_{0nn}^0 = 400$; (c,d) $\Delta_2 = 0$. All other parameters are the same as in Fig. 2.

caused by the interference of two-step and two-photon processes. The plots indicate that, due to the time-dependent shift of one-photon resonance induced by the second field, the dynamics of photodissociation and its dependence on the pulse sequences and shift changes considerably with the increase of the peak intensity of the second field [plots (a) and (b)]. At a given pulse-shift there exists an optimal intensity of this field [plot (c)]. The increase of the intensity of the second field leads to decoupling of the first one with the transition $mn$ [plot (d)] and, consequently, to a decrease in the overall dissociation [plot (c)]. Rabi oscillations of the populations at the transition $mn$ in the range of relatively small intensities of the second field [plot (d)] may give rise to substantial modification of the dynamics and to suppression of dissociation [plot (b)].
FIG. 5: Dissociation and population of levels driven by three laser pulses. \( \Omega_{mn} = \Omega_{nf} = 0, \ g_{mn}^0 = 2, \ g_{nm}^0 = 0.25, \ g_{ff}^0 = 0.36, \ g_{nf}^0 = 0.3, \ d_2 = 1, \ d_3 = 1.6; \Delta_3 = 0. \) The Fano parameters are \( q_{mn} = 0.2, \ q_{ff} = -0.5, \ q_{nf} = 10. \) Initially, only level \( m \) is populated. (a) and (b) – dependence on the pulse delay. (c) and (d) – dynamics of the populations and dissociation \( (\Sigma |a_j|^2 \) is the sum of populations including the energy integrated population of the continuum \( W)\). (c) \( \Delta_2 = 0, \) (d) \( \Delta_2 = -1.5. \)
FIG. 6: Dependence of the three-pulse driven population of level $f$ on the Fano parameter $q_{nf}$ (a) and on the two-photon detuning $\Delta_{nf}$ (b). $\Delta_2 = 2.8$, $\Delta_3 = 0$. (a) $\Omega_{nf} = 0$, (b) $q_{nf} = 10$. All other parameters are the same as in the previous figure.

FIG. 7: Dependence of the dissociation and of the population of the intermediate level $n$ on the two-photon detuning, where $q_{nf} = 10$. All other parameters are the same as in the previous figure.

C. Three-pulse control of photodissociation and population transfer by continuum coherence

With an understanding of the features described above, we shall investigate the dynamics of more complex scheme which embraces both schemes considered above by assuming all three fields at $\omega_1$, $\omega_2$ and $\omega_3$ (Fig. 1) turned on. First, we shall simulate the case when one-,
two- and three-photon resonances are fulfilled ($\Omega_{mn} = \Omega_{nf} = 0$), and the first and second pulses are peaked at the same instant as the third pulse, which is somewhat longer. The second pulse is of the same duration as the first one and can be either advanced or delayed with respect to the other two.

Figure 5 [plots (a) and (b)] shows that the dissociation output and population of the upper level can be controlled within the wide intervals by adjustment of the sequence and the time shift of the second pulse. With two examples [plots (c) and (d)], the simulation demonstrates the feasibility of achieving either comparable populations of all three discrete and integrated continuum states by the end of the pulses [plots (c), where all pulses peak at the same instant] or, alternatively, of ensuring a dominant population of the upper discrete state and dissociation output by advancing the second pulse regarding the other two [plot (d)]. Here, $\Sigma |a_j|^2$ is the sum of populations including the energy integrated population of the continuum $W$. Figure 6 [plot (a)] shows that the dynamics of the populations is considerably different for different Fano parameters (keeping the other parameters the same). It also shows that, for fixed time delays, the output can be controlled by the adjustment of appropriate two-photon detuning [plot (b)]. Figure 7 shows similar feasibilities of manipulation of the populations of the intermediate level and dissociation. Dark resonances, characterized by suppressed dissociation and selective population redistribution between the coupled bound states, or, alternatively, enhanced dissociation are shown to be achieved by variation of two-photon (and consequently three-photon) detuning.

IV. CONCLUSION

The theory of adiabatic passage between three discrete levels and a low-lying dissociation continuum is developed. Possible quantum control of two-photon dissociation ($\Lambda$-scheme) using auxiliary short laser pulses at adjacent bound-free transitions ($V$-configuration) is shown. The proposed three-pulse scheme enables one to manipulate dissociation through the dark states not connected with the ground one by the allowed transition. Besides dissociation, the scheme under investigation enables one to control the population transfer between two upper discrete levels via the lower-energy dissociation continuum, while a direct transition between these states is not allowed. A system of coupled equations for the probability amplitudes is employed, where three time-shifted pulses drive strongly the quantum system, so
that nonlinear quantum interference occurs through the overlap of two laser-induced continuum structures. The opportunities of manipulating photoinduced processes by judiciously frequency-detuned and time-delayed pulses are explored through extensive numerical simulations with the aid of a model relevant to typical experiments. The simulations display good agreement with the known experiments on two-pulse population transfer between metastable states via the ionization continuum and with experiments on two-photon photodissociation. The dependence of the dissociation spectrum on the magnitudes of the composite Fano parameters for excited levels, on detuning between two laser-induced continuum structures, on intensities, and on the counterintuitive order and delay of the pulses are investigated. We have demonstrated the opportunities of considerable suppression of photodissociation and considerable transfer of the lower-state population to the excited states or, alternatively, almost complete photodissociation, or redistribution the population between continuous and discrete states with the required ratio by the end of the pulses. The gained efficiency and flexibility may compensate for the increase in experimental complexity.

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