Nonadiabatic optical transitions as a turn-on switch for pulse shaping

F. A. Hashmi*
Centre for Quantum Physics, COMSATS Institute of Information Technology, Islamabad, Pakistan

M. A. Bouchene
Laboratoire de Collisions Agrégats Réactivité, CNRS UMR 5589, IRSAMC Université Paul Sabatier, 118 Route de Narbonne, 31062 Toulouse Cedex 4, France
(Received 28 June 2010; published 26 October 2010)

A strong nonresonant, asymmetric ultrashort pulse drives an atomic transition and causes a complete population inversion because of a sudden nonadiabatic jump. This jump is probed in real time by propagating a weak ultrashort pulse in the system which is resonant on an adjacent transition. The probe at the exit of the medium presents an oscillatory structure with the nonadiabatic jump marked in time by the onset of oscillations. The nonadiabatic jump thus acts as a "turn-on" switch for the shaping of the probe.

DOI: 10.1103/PhysRevA.82.043432 PACS number(s): 42.50.Md, 31.50.Gh, 42.65.Re, 42.50.Hz

I. INTRODUCTION

Atomic systems interacting with strong ultrashort pulses give rise to a lot of interesting phenomena that can often be well explained in the framework of the adiabatic basis [1]. The adiabatic basis presents light-shifted energy levels and thus explains the phenomena arising because of these modified energy levels. Such phenomena include, for example, transition probabilities in adiabatic following [2], rapid adiabatic passage [3,4], and the generation of new frequency components in the system [5]. Light shifts can also be combined with the dispersion effects of propagation and can be used for shaping purposes. A weak probe propagating in a light-shifted medium develops an oscillatory structure with oscillations at a time scale much shorter than the pulse width [5,6]. A second feature of the adiabatic basis is nonadiabatic transitions [7] between light-shifted energy levels, and these transitions depend strongly on the shape of the pulse [8]. By suitably shaped pulses, these nonadiabatic transitions can be significantly enhanced and an almost complete population transfer by an asymmetric pulse can be realized [9–11]. The complete population transfer in these cases is attributed to a sudden nonadiabatic jump in adiabatic population that arises because of a π area δ-function-like nonadiabatic coupling. In a previous study [12] a scheme to realize coherent control of such a nonadiabatic jump, and consequently control of the transition probability, has been proposed. In the present contribution we focus on the observation of the jump in the temporal profile of a field, and the use of such a jump for shaping purposes. Indeed the jump takes place in adiabatic dressed states and the observation of the jump in bare-state atomic populations through fluorescence spectroscopy is not straightforward [9]. We show here that this jump can be rendered observable by making use of propagation effects and can be used as a "turn-on" switch for pulse shaping.

This paper is organized as follows. In Sec. II we present the system. In Sec. III we recall the behavior of nonadiabatic transitions induced by a strong, nonresonant asymmetric pulse. In the last Sec. IV we present the use of propagation effects to probe the nonadiabatic jump and the use of the nonadiabatic jump as a control of pulse shaping. Finally we conclude.

II. SYSTEM

Consider a three-level atomic system in Λ configuration interacting with a strong "control" and a weak "probe" ultrashort field on different transitions as shown in Fig. 1. The control field is nonresonant on the $|a\rangle \leftrightarrow |c\rangle$ transition and it induces nonadiabatic effects on this transition. These effects take the form of a sudden, abrupt jump and the jump is probed by the weak probe field that propagates in the system and that is resonant on the $|b\rangle \leftrightarrow |c\rangle$ transition. The expressions for the two fields are $\vec{e}_z A_c f_c(t,y) e^{-i(\omega_c t - ky)}$ + c.c. (control) and $\vec{e}_z A_p f_p(t,y) e^{-i(\omega_{bc} t - ky)}$ + c.c. (probe) where A_i $(i = c,p)$ are the amplitudes of the two fields and f_i are the field envelopes normalized to unity at the entrance of the medium (at $y = 0$). The two fields are linearly polarized along the z axis and propagate along the y axis. We introduce dimensionless time and space variables as $T = (t - y/c)/\tau_c$ and $Y = y/L$ where τ_c is the characteristic time that will be defined later and L is the length of the medium. We further define dimensionless strength parameters for the two fields as $\theta_c = D_{ac} A_c \tau_c/\hbar$ and $\theta_p = D_{bc} A_p \tau_c/\hbar$ with D_{ac} and D_{bc} being the dipole matrix elements for the respective transitions, and the detuning of the control field as $\Delta = (\omega_{ac} - \omega_c)\tau_c$. The time evolution of the wave function

$$|\psi\rangle(T,Y) = a(T,Y)|a\rangle + b(T,Y)e^{-i(\omega_c - \omega_{bc})\tau_c T}|b\rangle + c(T,Y)e^{-i\omega_c \tau_c T}|c\rangle, \quad (1)$$

within the rotating wave approximation is given by

$$i\partial_T \begin{pmatrix} a \\ b \\ c \end{pmatrix} = \begin{pmatrix} 0 & 0 & -\theta_c f_c^* \\ 0 & \Delta & -\theta_p f_p^* \\ -\theta_c f_c & -\theta_p f_p & \Delta \end{pmatrix} \begin{pmatrix} a \\ b \\ c \end{pmatrix}. \quad (2)$$

The strongly driven transition ($|a\rangle \leftrightarrow |c\rangle$) can be transformed into the adiabatic basis defined as

$$\begin{pmatrix} |\alpha\rangle \\ |\gamma\rangle \end{pmatrix} = \begin{pmatrix} \cos(\theta) & \sin(\theta) \\ -\sin(\theta) & \cos(\theta) \end{pmatrix} \begin{pmatrix} |a\rangle \\ e^{-i\omega_c \tau_c T}|c\rangle \end{pmatrix}, \quad (3)$$

*faheel4@comsats.edu.pk

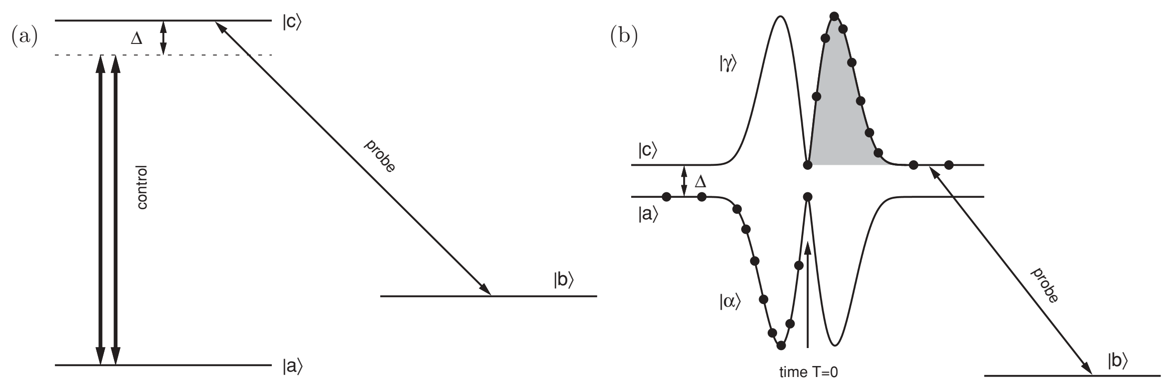

FIG. 1. (a) System in bare states (without RWA) and (b) System in dressed state (with RWA). The states $|a\rangle$ and $|c\rangle$ are light-shifted to $|\alpha\rangle$ and $|\gamma\rangle$ during the interaction. Small circles show time evolution of the system with a sudden nonadiabatic jump at $T=0$. The shaded region corresponds to the frequencies involved in the shaping of the probe. The probe effectively couples the state $|b\rangle$ with the state $|\gamma\rangle$ only and for the times when the light shifts are at minimum.

where θ is defined at the entrance of the medium (at $Y=0$) as $\tan 2\theta(T) = 2\theta_c f_c(T,0)/\Delta$. The time evolution of the dressed wave function

$$|\psi_d\rangle(T,Y) = \alpha(T,Y)|\alpha\rangle + \beta(T,Y)|b\rangle + \gamma(T,Y)|\gamma\rangle, \quad (4)$$

is given by

$$i\partial_T \begin{pmatrix} \alpha \\ \beta \\ \gamma \end{pmatrix} = \begin{pmatrix} \frac{\Delta-\Omega}{2} & -\theta_p f_p \sin(\theta) & i\partial_T\theta \\ -\theta_p f_p^* \sin(\theta) & \Delta & -\theta_p f_p^* \cos(\theta) \\ -i\partial_T\theta & -\theta_p f_p \cos(\theta) & \frac{\Delta+\Omega}{2} \end{pmatrix} \begin{pmatrix} \alpha \\ \beta \\ \gamma \end{pmatrix}. \quad (5)$$

Here $\Omega(T) = \sqrt{\Delta^2 + 4\theta_c^2 f_c^2(T,0)}$ is the light shift induced by the driving pulse and $\partial_T\theta$ is the nonadiabatic coupling between these light-shifted energy levels.

III. NONADIABATIC JUMP FOR STRONG DRIVING PULSE

We first consider the case where the probe is absent and the transition $|a\rangle \leftrightarrow |c\rangle$ is strongly driven by the nonresonant *zero area* asymmetric control field. The expression for the envelope of the control is

$$f_c(T,0) = \frac{1}{\sqrt{\pi}}\left[e^{-(T-\frac{1}{2})^2} - e^{-(T+\frac{1}{2})^2}\right]. \quad (6)$$

The envelope consists of two Gaussians that are time and phase delayed by τ_c and π, respectively, and where each Gaussian has the temporal width τ_c. This can arise, for example, in interferometers where a single Gaussian beam is split by a beam splitter, and the components travel along different paths to achieve the required time and phase delays. We are interested in the population dynamics and the asymptotic population transfer to the excited state by such excitation. Figure 2 shows the population profile of the excited state. We see that we have an almost complete population transfer by the control field which is counterintuitive as the control has no resonant components and has *zero* pulse area. The result can be explained in terms of the nonadiabatic jump (NAJ) that is a sudden jump in the dressed-state population occurring at $T=0$ when the control envelope has the maximum slope [9]. Indeed the coupling between the dressed states approximates to the π area δ function in the limit of strong driving and explains the population transfer [12]. This NAJ takes place in adiabatic populations, and observing it in the bare-state population is not obvious.

We see in Fig. 2 that the bare-state population shows strong oscillations. The oscillations arise at the onset of the excitation

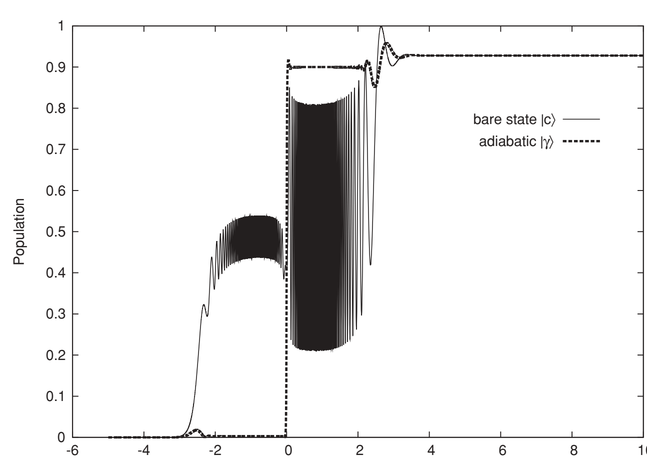

FIG. 2. Population dynamics for excited state in bare (solid) and dressed (dashed) states. A sudden nonadiabatic jump at $T=0$ transfers almost all of the population to the excited state. The jump is revealed in the bare-state population only by a change in the oscillation amplitude. Parameters are $\theta_c = 400$, $\theta_p = 0$, and $\Delta = 8$.

signifying that there is some population transfer to the state $|\gamma\rangle$ before $T = 0$. Indeed the nonadiabatic coupling follows the time derivative of the field amplitude and hence is nonzero at the onset of the excitation. Moreover, at the same time the induced light shifts are small. Hence a little population is transferred nonadiabatically to the excited adiabatic state, as can be verified by a small bump in the adiabatic population in Fig. 2. The peak of the bump at $T \approx -2.5\tau_c$ corresponds to 1.7% population, which subsequently falls to 0.2%. However, even this small population interferes with the population in the state $|\alpha\rangle$ and results in a 10% oscillation amplitude for the population in state $|c\rangle$ as we see in the figure. At the onset of the jump, the oscillation amplitude of the bare-state population changes, corresponding to the change of populations in adiabatic levels. However, the change in adiabatic populations is not always accompanied by observable effects in bare-state populations. Indeed, in the case of an *ideal* $0 \rightarrow 1$ jump in adiabatic populations at $T = 0$, there is no population transfer before $T = 0$, and the oscillations in the bare state completely disappear. In this case the signature of the nonadiabatic jump is completely lost in the bare-state populations and in the fluorescence signal from the system. We next propose a scheme where we can detect such nonadiabatic jumps.

IV. PROBING NONADIABATIC JUMP BY WEAK PROPAGATING PROBE PULSE

We next introduce the weak probe field in the system that is resonant on the $|b\rangle \leftrightarrow |c\rangle$ transition. The envelope of the probe at $Y = 0$ is given by

$$f_p(T,0) = \frac{1}{\sqrt{\pi}} e^{-\left(\frac{T-T_d}{\tau_p}\right)^2}. \tag{7}$$

Here T_d (in units of τ_c) is the dimensionless delay between the control and the probe field and τ_p (in units of τ_c) is the dimensionless temporal width of the probe. τ_p^{-1} is therefore the spectral width of the probe field. In the adiabatic picture the probe couples both $|\alpha\rangle$ and $|\gamma\rangle$ with the state $|b\rangle$; however, for $\Delta \gg \tau_p^{-1}$ the probe is resonant only with $|\gamma\rangle$. We next consider the propagation of the probe inside the medium and work out an analytical expression for the transmitted probe intensity. The probe field obeys the propagation equation [13]

$$\partial_Y f_p = i \frac{e_{\text{disp}}}{\theta_p} \beta^* (\alpha \sin\theta + \gamma \cos\theta), \tag{8}$$

where $e_{\text{disp}} = N D_{bc}^2 \omega_{bc} L \tau_c / (2c\epsilon_0 \hbar)$ (with N the atomic density) is the dispersion parameter that characterizes the severity of the propagation effects on the transmitted probe. The control field also obeys a similar equation of propagation but is only slightly distorted for $\theta_c \gg e_{\text{disp}}$. A detailed discussion of this dispersion parameter can be found in [14]. A perturbative solution of Eq. (8) in the limit of the small dispersion parameter ($e_{\text{disp}} < 1$) and with $\Delta \gg \tau_p^{-1}$ can be written as

$$f_p(T,Y) \approx f_p(T,0) + i e_{\text{disp}} \beta^*(T,0)\, \gamma(T,0) \cos(\theta)(T,0). \tag{9}$$

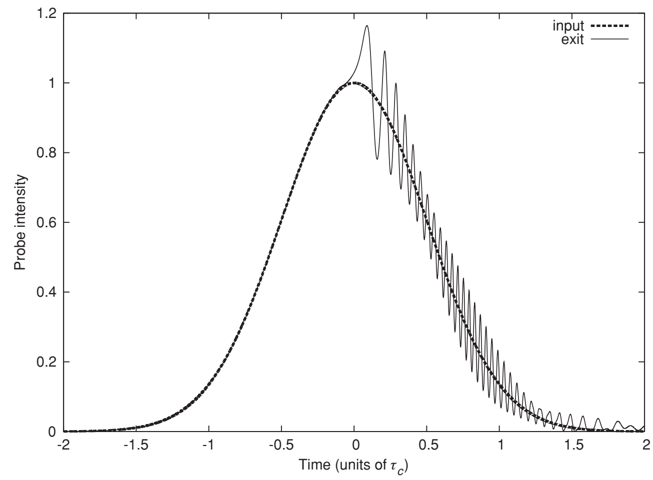

FIG. 3. Normalized probe intensity at input (dashed) and exit (solid) of the medium. At exit the probe develops an oscillatory structure with oscillations starting at the time of the nonadiabatic jump. Parameters are $\theta_c = 400$, $\theta_p = 0.1$, $\Delta = 8$, $T_d = 0$, and $e_{\text{disp}} = 1$.

We next work out the approximate solution for the above equation and consider the time evolution of the system. With initially all the population in the ground state $|\alpha\rangle$, and with adiabatic evolution, the population rests in the ground state until $T = 0$ as is shown in Figs. 1 and 2. Up to this point in time, the second term on the right-hand side of Eq. (9) vanishes and the probe propagates in the medium without any modifications as can be seen in Fig. 3. After $T = 0$, the probe develops an oscillatory structure which can be understood as follows. At $T = 0$, the nonadiabatic jump takes place and the excited state $|\gamma\rangle$ is suddenly populated. The subsequent evolution of the system for $T > 0$ and at *zeroth* order with respect to the probe amplitude is given by

$$\alpha^{(0)}(T) = 0, \tag{10}$$

$$\gamma^{(0)}(T) = e^{-i\int_0^T \frac{\Delta+\Omega(T')}{2} dT'}. \tag{11}$$

At *first* order, the amplitude β evolves for $T > 0$ as

$$\beta^{(1)}(T) = i\theta_p e^{-i\Delta T} \int_0^T f_p^*(T') \cos\theta\, e^{i\Delta T'} \gamma^{(0)}(T') dT'. \tag{12}$$

We further distinguish between the resonant and nonresonant contributions to $\beta^{(1)}$. The resonant contribution is for the time between $-T_r$ and T_r, which are the solutions of $[\Omega(T) - \Delta] \approx \tau_p^{-1}$. This is the time where the light shifts are insignificant and the excitation can be considered resonant. Outside these limits, the important light shifts stretch the levels apart and make the probe nonresonant. For the resonant contribution we make the approximation $\Omega(T) \rightarrow \Delta$ and $\cos\theta \rightarrow 1$. Equation (12) thus simplifies for $0 < T \leqslant T_r$ as

$$\beta^{(1)}(T) = i\theta_p e^{-i\Delta T} \int_0^T f_p^*(T') dT'. \tag{13}$$

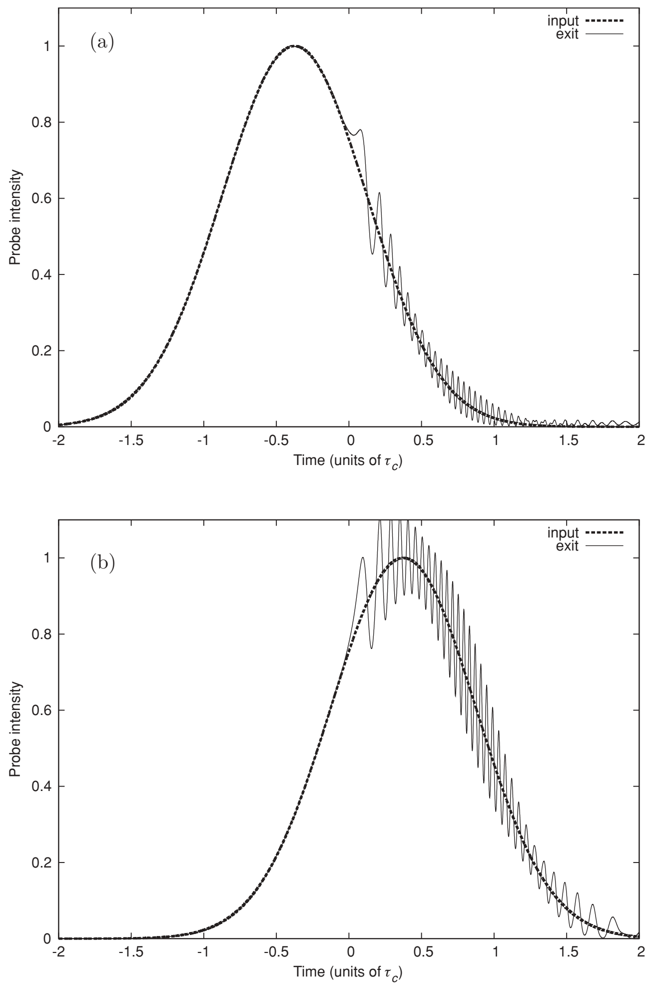

FIG. 4. The probe is (a) forward in time and (b) delayed in time with respect to the nonadiabatic jump. The oscillations always start at the onset of the jump at ($T = 0$). Parameters are (a) $T_d = -0.375$ and (b) $T_d = 0.375$. The other parameters are the same as in Fig. 3.

For subsequent times ($T > T_r$) the probe becomes nonresonant and the amplitude $\beta^{(1)}$ is given by

$$\beta^{(1)}(T) = i\theta_p e^{-i\Delta T} \int_0^{T_r} f_p^*(T')dT'. \qquad (14)$$

Finally using Eqs. (11) and (14) in Eq. (9), we can write the envelope of the field inside the medium as

$$f_p(T,Y) \approx f_p(T,0) + g(T,0)e^{-i\int_0^T \frac{\Omega(T')-\Delta}{2}dT'}, \qquad (15)$$

with $g(T,0) = e_{\text{disp}} \cos\theta(T,0) \int_0^{T_r} f_p(T',0)dT'$. The transmitted intensity $I_p(T,Y) = |A_p f_p(T,Y)|^2$ can thus be approximated as

$$I_p(T,1) \approx I_p(T,0) + 2A_p^2 f_p(T,0)g(T,0) \times \cos\left(\int_0^T \frac{\Omega(T')-\Delta}{2}dT'\right). \qquad (16)$$

The above expression clearly shows that the probe develops an oscillatory structure with the oscillation frequency given by the integral of the light-shifted region. This region is shown as shaded in Fig. 1(b) The oscillations can be seen in Fig. 3, and the onset of the oscillations marks the nonadiabatic jump in real time. In this manner propagating effects can be used to probe the nonadiabatic jump. Alternatively, we can look at this phenomenon as that of pulse shaping and can regard the nonadiabatic jump as the turn-on switch for shaping. The oscillations that appear on the temporal profile of the probe are at a time scale much shorter than the pulse width. These oscillations can be enhanced in amplitude by increasing the dispersion parameter. Also it should be noted that these oscillations arise because of the new frequency components that have been added in the probe spectrum [5], and which are in contrast with the standard passive pulse shaping techniques.

We further demonstrate our point in Fig. 4 where we introduce a delay between the probe and the control fields. The nonadiabatic jump still takes place at $T = 0$, but the probe is either forwarded or delayed in time. This causes different regions of the probe to be shaped with the onset of shaping still marked by the nonadiabatic jump at $T = 0$.

In conclusion, we have shown that the effects induced by an asymmetric shaped, strong, and nonresonant pulse on an atomic transition can lead to spectacular modifications in the temporal profile of a weak probe that acts resonantly on an adjacent transition. Nonadiabatic coupling between the driven transition brings, in a sudden manner, all the population to the adiabatic excited state. This jump, masked in strong oscillations in the bare-state population, can be rendered observable in the temporal profile of the probe that propagates in the medium. At the time of the jump the probe establishes a coherence and subsequent radiation of the atoms during the light-shifted region induces oscillations on the temporal profile of the probe. The scheme can thus be used for wave-shaping purposes and in this case the nonadiabatic jump acts as a turn-on switch for the shaping process. In slightly different excitation schemes this nonadiabatic jump can also be made to act as a turn-off switch for pulse shaping. For example, if initially all the population is in the excited state and the temporal delay between the fields is appropriate, then the modulations start before the nonadiabatic jump, and (in case of a perfect $0 \to 1$ jump) die out at the onset of the jump. Similarly in a three-level system in the V configuration a perfect $0 \to 1$ jump can act as a turn-off switch for the shaping of the probe. This type of pulse shaping, although limited in terms of the complexity of the shapes that can be realized, nevertheless has several advantages over traditional pulse shapers. For example, it can work both in the ultraviolet and long-pulse regimes (nanosecond scale). For femtosecond pulses, the oscillations can be in the attosecond range and this technique can provide a new route to produce attosecond pulses as long as the rotating wave approximation and the three-level model are preserved.

ACKNOWLEDGMENT

F.A.H. sincerely acknowledges the helpful discussion with M. S. Zubairy.

[1] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Atom-Photon Interactions: Basic Processes and Applications* (Wiley, New York, 1998).

[2] M. D. Crisp, Phys. Rev. A **8**, 2128 (1973).

[3] C. Liedenbaum, S. Stolte, and J. Reuss, Phys. Rep. **178**, 1 (1989).

[4] L. Allen and J. H. Eberly, *Optical Resonance and Two Level Atoms* (Dover, New York, 1975).

[5] J. C. Delagnes, F. A. Hashmi, and M. A. Bouchene, Phys. Rev. A **74**, 053822 (2006).

[6] J. C. Delagnes and M. A. Bouchene, Phys. Rev. A **69**, 063813 (2004).

[7] B. W. Shore, *The Theory of Coherent Atomic Excitation* (Wiley, New York, 1990).

[8] P. R. Berman, L. Yan, K.-H. Chiam, and R. Sung, Phys. Rev. A **57**, 79 (1998).

[9] G. S. Vasilev and N. V. Vitanov, Phys. Rev. A **73**, 023416 (2006).

[10] N. V. Vitanov, New J. Phys. **9**, 58 (2007).

[11] B. T. Torosov and N. V. Vitanov, Phys. Rev. A **76**, 053404 (2007).

[12] F. A. Hashmi and M. A. Bouchene, Phys. Rev. A **79**, 025401 (2009).

[13] P. W. Milonni and J. H. Eberly, *Lasers* (Wiley, New York, 1988).

[14] J. C. Delagnes and M. A. Bouchene, Opt. Commun. **281**, 5824 (2008).