Phase control of nonadiabatic optical transitions

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We theoretically study the interaction of two time delayed, phase-locked, and nonresonant pulses with a two-level system in the strong field regime. The population transfer is shown to be extremely sensitive to the phase shift ϕ between the pulses, with efficient population transfer taking place only for ϕ close to π. This effect is explained in terms of nonadiabatic jump and rapid adiabatic passage phenomena.

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The interaction of strong fields with quantum systems leads to a variety of phenomena that have no equivalent in the weak field regime. Of particular interest are nonadiabatic transitions between induced adiabatic energy levels when strong nonresonant light pulses interact with atomic systems [1]. They can lead to significant asymptotic population transfer to the excited bare state even when the pulse spectrum does not cover the atomic transition. This is in complete contrast with the simplified version of light matter interaction, in which, a photon can only be absorbed if the resonance condition is satisfied. This simplified version turns out to be true only in the weak field regime. Strong pulses introduce adiabatic energy levels in the system and nonadiabatic transitions between these energy levels are reflected in asymptotic bare state populations. The excitation can cause level crossings, in which case adiabatic rapid passage (ARP) comes into action and leads to a complete inversion of population in the system [2–4]. This phenomenon is efficient and robust against laser parameters. Situations where level crossings do not appear have also been investigated. For symmetric strong pulses, it has been shown that the population transfer is extremely dependent on the shape of the pulse [5] through nonadiabatic coupling. If asymmetric pulses are used (which present phase jump or a change of sign for the amplitude), the nonadiabatic coupling can be so significant that complete population transfer can be realized [6–8]. This result is especially striking when the fields have zero pulse area. Resonant excitation with zero area pulse leads to no population transfer whereas nonresonant excitation can lead to complete population inversion [6].

In this paper, we study the population transfer induced by a sequence of two time delayed, identical, and phase locked pulses interacting nonresonantly with an atomic system. For the relative phase shift $\phi=\pi$ between the two pulses, the total pulse area is zero and nonadiabatic jump (NAJ) leads to an almost complete population transfer as already studied by Vitanov and collaborators in detail in [6]. The originality in the present study is the very sensitive phase dependence of nonadiabatic coupling and is a consequence of the high nonlinearity of the interaction. The population transfer efficiency and asymptotic population in the excited state are severely affected when the phase shift moves slightly away from π, whereas for large deviation from π, a double ARP effect is responsible for vanishing population transfer. This results in a very narrow peak in asymptotic population in the excited state as the function of relative phase shift between the two pulses, as shown in Fig. 3. With the virtue of this increased sensitivity, this effect can lead to the improvement of the techniques based on interferometry such as lock-in techniques, metrology, Ramsey spectroscopy, coherent control, pump-probe techniques, and gyrolaser techniques to name some [9–11].

We consider the two-level system with states $|a\rangle$ and $|b\rangle$ (energies 0 and $\hbar\omega_0$, respectively) with initially all the population in the ground state $|a\rangle$. The system is driven by a strong ultrashort field $E_d(t)=\epsilon_{0d}f_d(t)e^{-i\omega_d t}+\text{c.c.}$ where ϵ_{0d} is the field amplitude, $f_d(t)$ is the envelope of the pulse, and c.c. stands for complex conjugate. The envelope $f_d(t)$ consists of two time delayed Gaussians each having the time duration τ_d. The delay between the two Gaussians is τ (in units of τ_d) and the two are dephased by the relative phase difference ϕ. In dimensionless time $T=t/\tau_d$, the envelope is given by

$$f_d(\phi,T)=\frac{1}{\sqrt{\pi}}\left(e^{-(T+\tau/2)^2}+e^{i\phi}e^{-(T-\tau/2)^2}\right). \tag{1}$$

The central frequency of the pulse ω_d is detuned from the resonance ω_0 and we define dimensionless detuning as $\Delta=(\omega_0-\omega_d)\tau_d$. The wave function of the system can be written as

$$|\Psi\rangle(\phi,T)=a(\phi,T)|a\rangle+b(\phi,T)e^{-i\omega_d\tau_d T}|b\rangle. \tag{2}$$

The Schrödinger equation with rotating wave approximation (RWA) leads to the following equations for the evolution of the system:

$$i\partial_T\begin{pmatrix}a\\b\end{pmatrix}(\phi,T)=\begin{pmatrix}0 & -\theta_d f_d^*\\ -\theta_d f_d & \Delta\end{pmatrix}\begin{pmatrix}a\\b\end{pmatrix}(\phi,T). \tag{3}$$

Here $\theta_d=\mu_{ab}\epsilon_0\tau_d/\hbar$ characterizes the strength of the driving field.

The interaction can be better studied by transforming the system into adiabatic basis. We define a rotation matrix as

$$R(T)=\begin{pmatrix}\cos\theta & \sin\theta\\ -\sin\theta & \cos\theta\end{pmatrix}(T), \tag{4}$$

where $\theta(T)$ is defined as

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$$\theta(T) = \frac{1}{2}\arctan[2rf_d(\pi,T)]. \tag{5}$$

Here $r=\theta_d/\Delta$ is an important parameter that characterizes the nonadiabatic coupling. The amplitudes of the wave function (2) are transformed in adiabatic basis as

$$\begin{pmatrix}\alpha_- \\ \alpha_+\end{pmatrix}(\phi,T) = R(T)\begin{pmatrix}a \\ b\end{pmatrix}(\phi,T). \tag{6}$$

By defining $R(T)$ at $\phi=\pi$, we contain all the phase dependence in the amplitudes (α_-,α_+). This (arbitrary) basis choice will prove to be pertinent in the present case, since the physics associated with the obtained results will be elegantly highlighted with this basis change. It should also be noted that as $R(T=\infty)=\left(\begin{smallmatrix}1 & 0\\ 0 & 1\end{smallmatrix}\right)$, the asymptotic bare state and adiabatic populations are the same ($|b|^2=|\alpha_+|^2$ at $T=\pm\infty$ for all ϕ). The time evolution of the amplitudes is given by

$$i\partial_T\begin{pmatrix}\alpha_- \\ \alpha_+\end{pmatrix}(\phi,T) = (A(T)+V(\phi,T))\begin{pmatrix}\alpha_- \\ \alpha_+\end{pmatrix}(\phi,T). \tag{7}$$

$A(T)$ represents the Hamiltonian for $\phi=\pi$. It is given by

$$A(T) = \begin{pmatrix}\dfrac{\Delta-\Omega(T)}{2} & i\partial_T\theta(T) \\ -i\partial_T\theta(T) & \dfrac{\Delta+\Omega(T)}{2}\end{pmatrix}. \tag{8}$$

The diagonal terms are the light shifted adiabatic energy levels with $\Omega(T)=\Delta\sqrt{1+4r^2f_d^2(\pi,T)}$ being the instantaneous separation between the levels, and the off-diagonal term $\partial_T\theta$ represents the nonadiabatic coupling. $V(\phi,T)$ in (7) represents the correction in energy levels and the coupling when $\phi\neq\pi$. It is given by

$$V(\phi,T) = \cos^2\frac{\phi}{2}\begin{pmatrix}\sin 2\theta & \cos 2\theta - i\tan\dfrac{\phi}{2} \\ \cos 2\theta + i\tan\dfrac{\phi}{2} & -\sin 2\theta\end{pmatrix}G(T) \tag{9}$$

with $G(T)=-2r\Delta e^{-[T-(\tau/2)]^2}/\sqrt{\pi}$. We next consider population dynamics for different values of ϕ.

For $\phi=\pi$, the two Gaussians are asymmetric with respect to each other, the matrix $V(\phi,T)=0$, and the dynamics is determined by the matrix $A(T)$ given in (8). For $|\tau|\gg\tau_d$ the two Gaussians dress the system independently from each other with each having local nonadiabatic coupling that can not cause significant population transfer. When the two Gaussians are brought near to each other (with $|\tau|\approx\tau_d$) such that the falling edge of one Gaussian coincides with the rising edge of the other, the light shifts and the nonadiabatic coupling add up nonlinearly. For $\tau=\tau_d$, the temporal profile of the field, the adiabatic energy levels, the nonadiabatic coupling, and the population in adiabatic excited state are shown in Fig. 1. Energy levels present a modulated structure with a node at $T=0$. The nonadiabatic coupling follows the derivative of the field profile and is given by

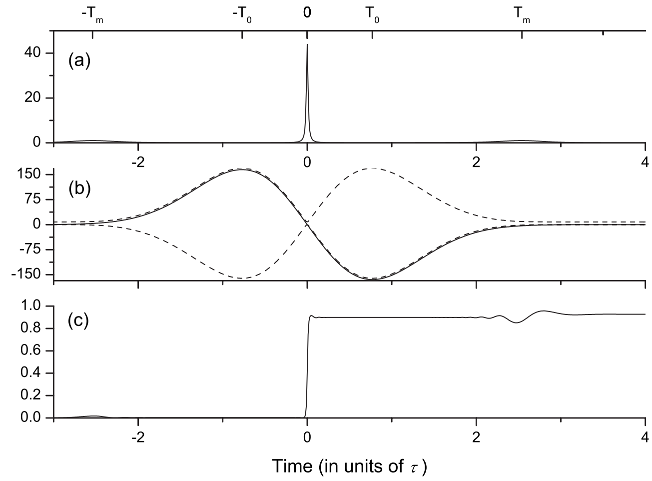

FIG. 1. (a) Nonadiabatic coupling $|\partial_T\theta|$; it presents a maximum at $T=0$, two local maxima at $T=\pm T_m$, and is *zero* at $T=\pm T_0$. (b) Field profile $f_d(\pi,T)$ (solid line) and adiabatic energy levels $\frac{\Delta\pm\Omega(T)}{2}$ (dashed line). (c) Excited adiabatic state population $|\alpha_+|^2$ showing NAJ. Parameters are $\Delta=8$, $\theta_d=400$ $(r=50)$, $\tau=1$, and $\phi=\pi$.

$$\partial_T\theta(T) = \frac{r\partial_T f_d(\pi,T)}{1+4r^2f_d^2(\pi,T)}. \tag{10}$$

Its absolute value $|\partial_T\theta|$ presents a maximum at $T=0$ where the light shifts vanish, and goes to zero at $\pm T_0$ [solution of $2T\tanh(\tau T)/\tau=1$] where the light shifts are at maximum. Two local maxima appear in the wings at $T=\pm T_m$.

Between $\pm T_0$ the nonadiabatic coupling behaves as a δ function for $r\to\infty$. Indeed, the field vanishes at $T=0$ $[f_d(\pi,0)=0]$, and the nonadiabatic coupling diverges for $r\to\infty$ $[\partial_T\theta(0)\to\infty]$. However, the area beneath the coupling $A_{NC}=2\int_{-T_0}^{T_0}(\partial_T\theta)(T)dT$ between $\pm T_0$ remains finite. It is given by $A_{NC}=2\arctan 2rf_d(\pi,T_0)$, and it behaves as $A_{NC}|_{r\to\infty}=-\pi$. The characteristic width of the coupling is $\delta T=A_{NC}/\partial_T\theta(0)$. It behaves as $\delta T=2\arctan 2rf_d(\pi,T_0)/r\partial_T f_d(\pi,0)$ and vanishes for $r\to\infty$. These results show that the central part of the nonadiabatic coupling around $T=0$ indeed behaves as a δ function with an area $-\pi$ (or π if the sequence of the two Gaussians is reversed). Moreover, near $T=0$, the light shifts are at minimum with $\Omega\approx\Delta$, and in the limit of strong pulse ($r\gg1$), the excitation can be considered as resonant. The transition probability to the excited adiabatic level is thus $\sin^2\frac{A_{NC}}{2}\approx1$. Complete population inversion in adiabatic basis with a sudden jump can thus be realized. This is shown in Fig. 1(c). A limitation to obtain a perfect 0 to 1 population jump in the adiabatic states is the population already transferred in the wing at $T=-T_m$. At $T=T_m$, the nonadiabatic coupling can again modify asymptotic population. This can be avoided if the evolution is adiabatic in the wings, requiring $\Omega(\pm T_m)\gg|\partial_T\theta(\pm T_m)|$. This condition can be easily fulfilled by making detuning large $\Delta\gg1$ and maintaining $r\gg1$ for obtaining the desired NAJ at $T=0$.

When $\phi\neq\pi$, additional contributions from the matrix $V(\phi,T)$ must be taken into account. For arbitrary times and phase shift values, V contains both nonvanishing diagonal

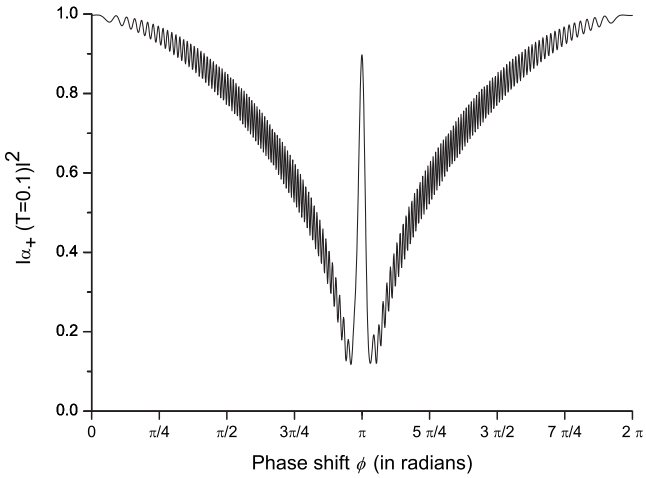

FIG. 2. Adiabatic excited state population profile at time ($T=0.1$) just after the nonadiabatic jump (for $\phi=\pi$). Other parameters are the same as those in Fig. 1.

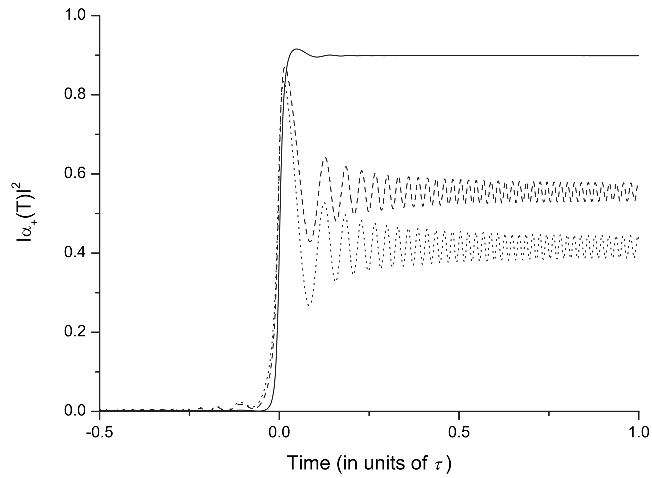

FIG. 4. Dynamics of adiabatic excited state population with $\phi=\pi$ (solid line), $\phi=\pi+0.04$ (dashed line), and $\phi=\pi+0.05$ (dotted line). Other parameters are the same as those in Fig. 1.

and off-diagonal terms. Light shifts and optical coupling between the adiabatic states are thus modified. As the total coupling is no longer a π area δ function, and the excitation is no longer resonant, the transition probability to excited adiabatic state is dramatically affected. In Fig. 2, the excited state adiabatic population as a function of ϕ is shown just after the time when a nonadiabatic jump occurs for $\phi=\pi$. It can be seen that close to $\phi=\pi$, the jump retains some of its character in a very narrow window while rapidly losing its efficiency. As we move away from the center, the population again continues to rise, but this latter rise is not due to NAJ. It is a transient phenomena caused by level crossings and no permanent population transfer takes place in the wings, as can be verified in the plot of asymptotic excited state population in Fig. 3. This latter demonstrates the sensitive dependence of transition probably on phase shift. The behavior around $\phi=\pi$ can be explained by analyzing the matrix elements of $V(\phi,T)$. For small variations of ϕ around π with $\phi=\pi+\epsilon$, $\epsilon \ll 1$, we can neglect the additional light shifts as these are proportional to ϵ^2. Only the coupling between the adiabatic states is modified. The matrix V at $T=0$ thus simplifies as $V \approx \frac{r\Delta\epsilon}{\sqrt{\pi}} e^{-\tau^2/4} \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}$. The modification of the transition probability follows a subtle scenario sketched in Fig. 4 that represents the transient dynamics for ϕ close to π. During the action of NAJ the modification of the effective pulse area due to the presence of the additional coupling V is negligible provided $2\left|\frac{r\Delta\epsilon}{\sqrt{\pi}} e^{-\tau^2/4} \delta T\right| \ll \pi$, i.e., $\epsilon \ll \frac{\tau}{\Delta}$ (δT is the time over which the NAJ transition occurs). In Fig. 4 we have $\frac{\tau}{\Delta}=0.125$. This explains why the maximum reached for $\phi=0.05$ and 0.04 is still important. Outside the interaction window δT, the nonadiabatic coupling $\partial_T \theta$ vanishes but $V(\phi,T)$ still acts. This leads to the modification of the population transfer for as long as the coupling $\frac{r\Delta\epsilon}{\sqrt{\pi}} e^{-\tau^2/4}$ is larger than the detuning Δ, i.e., $\epsilon > \frac{\sqrt{\pi} e^{\tau^2/4}}{r}$. This modification is in accordance with the rapid decrease of population observed in Fig. 4 and the sharp peak observed around $\phi=\pi$ in Figs. 2 and 3.

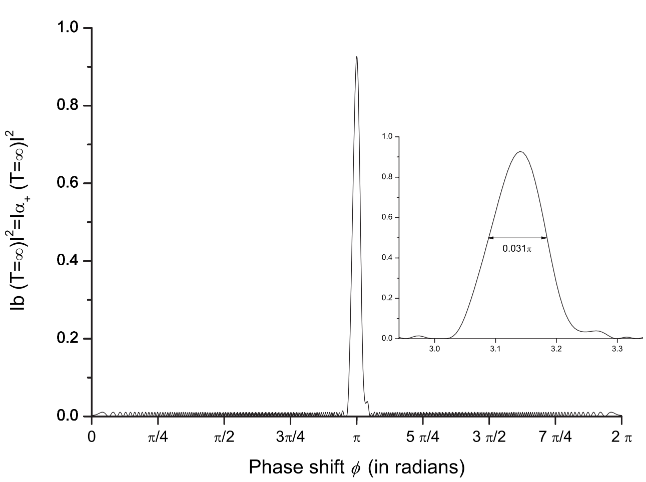

FIG. 3. Asymptotic excited state population. Parameters are the same as those in Fig. 1. In the inset a zoom is made to show the width of the peak.

An important feature is the presence of strong decaying oscillations in the population dynamics (for long times). These oscillations are the result of nonresonant contributions (because of important light shifts) that interfere with the resonant contribution. Although the nonresonant contributions to the population are small, they lead to observable effects because of the interference with the resonant contribution. These (ultrafast) coherent transients have been observed and reported in atomic systems driven by chirped pulses [12,13], or submitted to strong fields that induce light shifts [14–16].

The realization of very sharp structures with ϕ is in line with a very good spatial and temporal sensitivity of interferometers. For instance, when the sequence of two ultrashort pulses is obtained by sending a single pulse into an interferometer, we can write $\phi=\omega_d \delta t$, where δt is the delay between the two pulses. For pulses with 800 nm wavelength, we obtain from Fig. 3, $\delta t \sim 40$ as for field strength $\theta_d=400$. This corresponds to spatial resolution $\delta x = c\,\delta t$ of 12 nm.

Outside the peak for $\phi=\pi$, level crossings appear in the system. These arise because the diagonal element of V, $\sin 2\theta = 2rf_d(\pi,T)$ changes sign at $T=0$. Another crossing appears near the end of the driving field because adiabatic levels relax back to bare states. Any nonvanishing coupling at these crossings can cause significant (transient) population transfer in the adiabatic basis. This explains the transient adiabatic population in the wings at $T=0.1$ in Fig. 2. These crossings and the corresponding adiabatic population profile for $\phi=0$ is shown in Fig. 5. The transient oscillations are strongly attenuated in this case and only appear near the end of the pulse when the energy levels are close enough to cause nonresonant excitations.

We have shown that a sequence of two phase-locked strong pulses interacting nonresonantly with a two-level atomic system can lead to complete inversion of population. The phenomena occurs only for relative phase inside a small window around $\phi=\pi$. This shows the extreme dependence of the population transfer on phase, and it can render phase control of nonadiabatic transitions in the atomic system. The high degree of phase sensitivity can also be used to formulate new techniques for the stabilization of interferometers and paves the way for a very large range of applications. Attosecond resolution (corresponding to nanometer spatial resolution) can be reached in this way with strong pulses. Finally, a large value of θ_d can be obtained using long pulses.

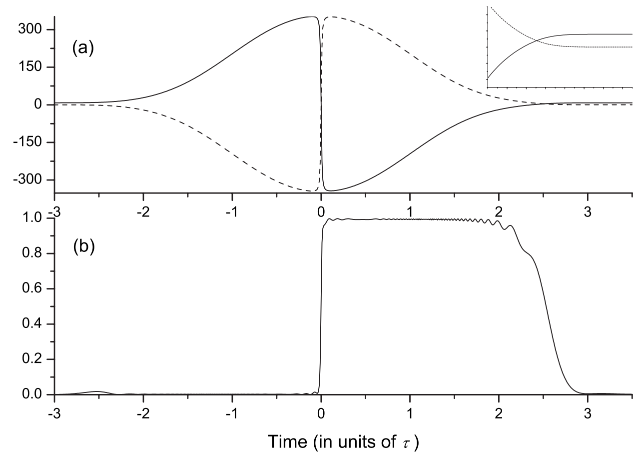

FIG. 5. (a) Modified adiabatic energy levels showing one crossing at $T=0$, and another near $T=2.5$; the second crossing is shown magnified in the inset. (b) Population in adiabatic excited state. The phase shift is $\phi=0$; other parameters are the same as those in Fig. 1.

For instance, we have $\theta_d \approx 110\mu_{ab}(\text{a.u.})\tau_d(\text{ns})\sqrt{I(\text{MW/cm}^2)}$. For a transition with $\mu_{ab}=4$ a.u., a laser pulse with a time duration $\tau_d=1$ ns, and an energy of 0.1 mJ focused on 1 mm^2 spot, we can obtain a value for θ_d as large as 440.

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