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Phase control of the temporal envelope of an ultrashort pulse propagating in an strongly driven atomic medium

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Abstract. We consider a double two-level system in which each single two-level system is driven by a strong linearly polarized pulse. The system is probed by a weak pulse orthogonally polarized. We study the distortion experienced by the probe temporal envelope when the relative phase between the pump and the probe is varied. We show that pulse behavior oscillates between two regimes that corresponds to transparency and amplification respectively. Interpretation of this effect is given within adiabatic representation.

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Key words: coherent control, propagation effects, light shifts

1 Introduction

The control of physical processes through manipulation of the relative phase between exciting fields represents the basic scheme of coherent control [1]. Several mechanisms in this regard have been explored. The combination of a fundamental frequency and its harmonics [2] and excitation by time delayed coherent pulses [3] are some examples. The control comes from the relative optical phase difference between different excitation fields. The attractive aspect in these experiments is the ease and the versatility in the control of this latter parameter. For example, for a sequence of pulses, it can be realized by modifying the relative delay between the pulses [4]. If strong pulses are used, light-shifts are induced in the medium and strong modification occurs in the atomic structure. Light shifts are the basis for many spectacular phenomena in atomic and molecular physics (rapid adiabatic passage, stimulated Raman adiabatic passage, Stark-chirped adiabatic passage, light induced potential etc. [5,6]). However, only little work deals with the control of these light shifts. An original situation

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arises in a double two-level system where each single two-level sub-system is driven by a strong pulse. We have shown in a series of papers [7–9] that coherent control ideas can be applied to the control of light-shifts induced by the strong field. An efficient control of the amplification of a weak pulse that probes the driven system is obtained. The scope of present paper is to investigate the temporal behavior of the probe pulse and observe its dependence with the phase and the intensity of the pump pulse. We demonstrate both experimentally and theoretically that depending on the relative phase between the pulses the probe pulse can be restored with little distortion (transparency regime) or with strong reshaping effects (amplification regime). We connect these features to the modification of the light shifts in the atomic sample for corresponding values of the relative phase.

2 Maxwell Bloch equations

We consider the situation of a duplicated two-level system $\{|1\rangle,|1'\rangle,|2\rangle,|2'\rangle\}$ (Fig. 1a) excited resonantly by two time delayed pulses. Pulse (1) strongly couples the parallel states while pulse (2) which is weak couples resonantly the crossed states. In practice, this situation can be realized by exciting the atomic rubidium with a pair of π- and σ-polarized pulses acting on the $S_{1/2}\rightarrow P_{1/2}$ transition. The electric fields of the pulses that propagate along the $\hat{y}$ axis are expressed as $\vec{E}_1(y,t)=\vec{e}_z(\epsilon_{01}f_1(y,t)e^{-i\omega t}+cc)$ and $\vec{E}_2(y,t)=\vec{e}_x(\epsilon_{02}f_2(y,t)e^{i\phi}e^{-i\omega t}+cc)$ with $f_1(y=0,t)=\pi^{-1/2}e^{-(t/\tau_0)^2}$, $f_2(y=0,t)=f_1(y=0,t-\tau)$, $\phi=\omega\tau$ is the relative phase between the pulses at the entrance of the medium, t represents the local time $(t=t_{lab}-y/c)$, τ is the delay and $|\epsilon_{02}/\epsilon_{01}|\ll 1$. Initially, the atoms are statistically equally distributed between the two ground states $|1\rangle$ and $|1'\rangle$. From symmetry arguments, we can consider the evolution of only those in state $|1\rangle$.

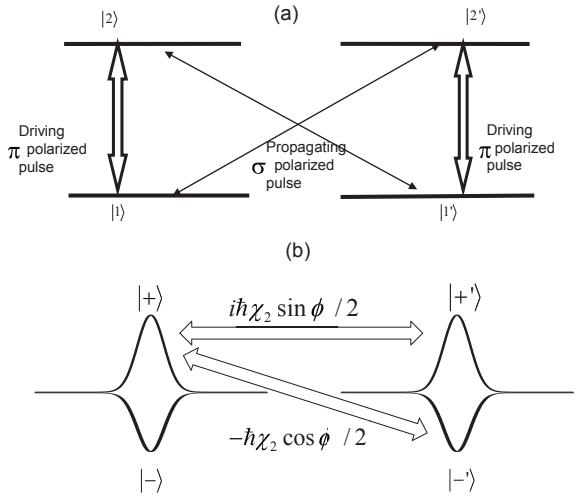

Figure 1: (a) Energy levels and optical transitions involved in our system. (b) Adiabatic representation (we represent only transitions connecting state $|+\rangle$).

The σ pulse is modified during propagation by both the dispersive effects due to the medium and the energy exchange with the driving pulse. Since the relaxation rates and the Doppler dephasing time are long here, the spectrum bandwidths of both σ and π-polarized pulses are much larger than the absorption line-width. The absorption from the medium is thus negligible. Moreover, for long times, all the excited atoms return back to the ground state emitting coherent radiation at a time scale much shorter than the excited states lifetime and the Doppler dephasing time. This is Free Induction Decay (FID). The absorption is thus only transient and there is no permanent deposition of energy in the medium. The σ pulse obeys the following equation of propagation [10]

$$\frac{\partial\left(f_2(Y,t)e^{i\phi}\right)}{\partial Y}=-i\frac{e_{disp}}{\theta_2}\rho^{(\sigma)}(Y,t) \tag{1}$$

with $Y=y/L$, L the length of the medium, $d=\langle 1|\vec{d}\cdot\vec{e}_x|2'\rangle$ is the dipole moment, $\theta_2=d\epsilon_{02}\tau_0/\hbar$ the pulse area of the σ pulse at the entrance of the medium (with $\theta_2\ll 1$), $e_{disp}=Nd^2\omega L\tau_0/(2c\epsilon_0\hbar)$ is a parameter that characterizes the severity of propagation effects, N is the atomic density, $\rho^{(\sigma)}=\rho_{2'1}+\rho_{21'}$, where $\rho_{ij}=\langle i|\rho|j\rangle$, $\rho=|\tilde{\psi}\rangle\langle\tilde{\psi}|$, $|\tilde{\psi}\rangle=e^{iH_0t/\hbar}|\psi\rangle$, H_0 is the Hamiltonian of the free atom and $|\psi\rangle$ the wavefunction. The driving pulses may also be modified during propagation and obeys to the following propagation equation

$$\frac{\partial\left(f_1(Y,t)\right)}{\partial Y}=-i\frac{e_{disp}}{\theta_1}\rho^{(\pi)}(Y,t) \tag{2}$$

with $\rho^{(\pi)}=\rho_{21}+\rho_{2'1'}$ and $\theta_1=d\epsilon_{01}\tau_0/\hbar$. The matrix density obeys to the time dependent differential equation

$$\frac{\partial}{\partial T}\rho=-\frac{i}{\hbar}[H,\rho] \tag{3}$$

with $T=t/\tau_0$ and

$$H=-\hbar\begin{pmatrix}0 & 0 & \theta_1 f_1^*(Y,T) & \theta_2 f_2^*(Y,T)e^{-i\phi}\\ 0 & 0 & \theta_2 f_2^*(Y,T)e^{-i\phi} & -\theta_1 f_1^*(Y,T)\\ \theta_1 f_1(Y,T) & \theta_2 f_2(Y,T)e^{i\phi} & 0 & 0\\ \theta_2 f_2(Y,T)e^{i\phi} & -\theta_1 f_1(Y,T) & 0 & 0\end{pmatrix}. \tag{4}$$

The set of Eqs. (1)–(3) describe completely the dynamics the combined field and atomic system. The phase dependence of these equations will reveal strong interference effects on the behaviour of the probe pulse at the exit of the sample that we describe experimentally next.

3 Experiment and results

Experimental observation of these effects has been achieved in rubidium on the $S_{1/2}\rightarrow P_{1/2}$ transition (794.7 nm). A regenerative amplifier pumped by a titanium sapphire laser delivers

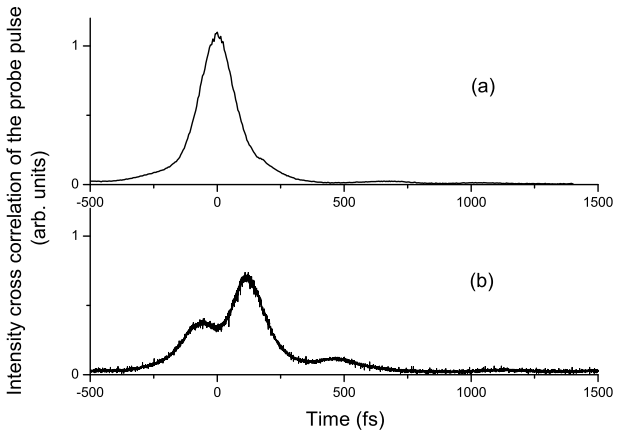

Figure 2: (a) Energy levels and optical transitions involved in our system. (b) Adiabatic representation (we represent only transitions connecting state $|+\rangle$).

linearly polarized laser pulses with 90 fs duration. They are split into two parts and recombined in a Mach-Zender interferometer, with a variable optical path difference resulting in a two-pulse sequence. In one arm of the interferometer a wave plate $\lambda/2$ combined with a polarizer rotates the polarization by 90° and allows also an eventual modification of the energy of the pulse. The beam emerging from this arm constitutes the driving π-polarized pulse whereas the σ-polarized pulse propagates in the other arm. The waist of π and σ-polarized pulses are 1.1 mm and 0.8 mm at the interaction region. The energy of the probe is 0.14μJ leading to a pulse area θ_2=0.1π . The heat pipe (length 12cm) was heated at a temperature T≈140°C for which e_{disp}≈0.2. The dimensionless absorption coefficient (optical depth) is $\alpha_0 L$≈8000≫1. At the exit, the two pulses are separated by a polarizer and the temporal profile of the weak transmitted pulse is measured through an intensity cross correlation with a reference pulse replica of the initial incident probe pulse. Experimental results are displayed in Figs. 2 and 3. Fig. 2 represents the temporal profile of the probe at the entrance of the medium (a) and at the exit (b) when no pump beam is applied. The probe pulse experiments in the atomic sample a strong distortion due to dispersion effects. When the pump pulse is applied, the behavior of the probe is modified depending strongly on the relative phase. In Fig. 3, we represent the temporal profile of the probe pulse for several intensities of the pump. Here, the phase stability was impossible to obtain. We turn this difficulty into an advantage by enhancing the phase fluctuations thanks to a vibrant device that allows the phase change when the delay between the reference and the probe is varied. When the delay between the reference and the probe is varied, the phase ϕ varies between 0 and $\pi/2$ as a consequence. Thus, rapid variations exhibited in Fig. 3 correspond to the phase modification when recording the signal. We distinguish easily between the envelopes corresponding to the cases $\phi = 0$ (dashed line) and $\phi = \pi/2$ (dotted line) and representing extreme situations (we represents in the figure the envelopes for the case corresponding to a pump energy about 150μJ). We see

that the signal oscillates between these two envelopes. One of these envelopes corresponds to a non distorted profile and the other one corresponds to a distorted signal with a long characteristic tail which amplitude increases when the pump energy is increased. The presence of the pump beam renders the atomic sample transparent or amplifier. We go next to the interpretation of these results.

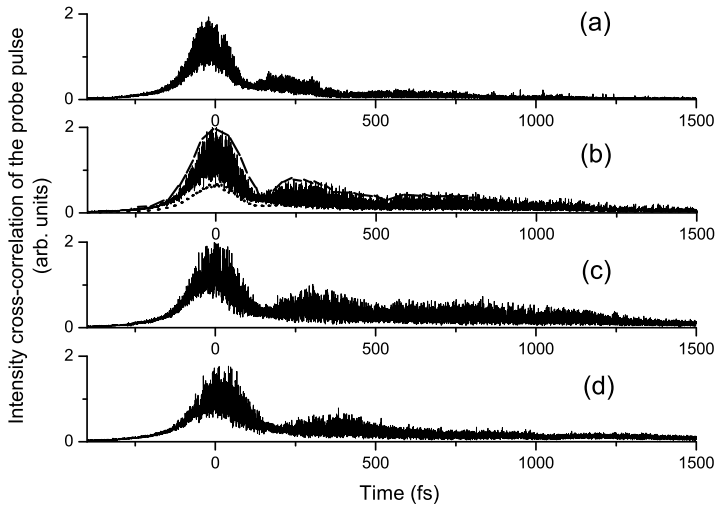

Figure 3: Experimental results. Temporal behaviour of the σ pulse for an atomic system driven by a strong field which energy is (a) 80 (b) 150 (c) 200 (d) 300 μJ. See text for experimental parameters. The phase fluctuation is responsible for the rapid variations.

4 Interpretation

The phase variation of the intensity signal reveals strong interference effects that take place inside the atomic system. A clear interpretation of the results can be obtained in the adiabatic picture, where the atomic system is dressed semi-classically. This description gives a complementary insight of the interaction and resorts the significance of the ϕ-dependence for the transmitted σ pulse. We define the adiabatic states by $|\pm\rangle = (e^{i\omega t}|2\rangle \mp |1\rangle)/\sqrt{2}$ and $|\pm'\rangle = (e^{i\omega t}|2'\rangle \mp |1'\rangle)/\sqrt{2}$. In this representation only cross states are coupled by the σ pulse (Fig. 1). The non-vanishing coupling elements are then

$$\langle -|H_\sigma|-'\rangle = -\langle +|H_\sigma|+'\rangle = -i\hbar\chi_2 \sin\frac{\phi}{2}, \tag{5a}$$

$$\langle +|H_\sigma|-'\rangle = -\langle -|H_\sigma|+'\rangle = -\hbar\chi_2 \cos\frac{\phi}{2} \tag{5b}$$

with $\chi_2 = d\epsilon_{02}f_2/\hbar$. The key point in this description is the distinction that appears between the parallel and anti-parallel adiabatic states, highlighting the ϕ-dependence. The parallel

states are coupled through the imaginary part of the σ pulse field — proportional to $\sin\phi$ — whereas the anti-parallel states are coupled through the real part — proportional to $\cos\psi$. The phase variation from $\pi/2$ to 0 leads to a progressive evolution from the situation where the interaction with the σ pulse is resonant to the case where it is not, performing a real control of light-shifts effects.

The interpretation of our experimental results can be highlighted within the adiabatic formalism. Indeed, for $\phi=0$ the probe σ pulse interacts only with anti-parallel states and the interaction becomes gradually non resonant as the light shift increases. For a sufficiently strong pump π pulse, the light shifts exceed the spectral width of the probe pulse and a transparency window for the σ pulse appears. The probe pulse is then almost unaffected by propagation and is transmitted with little modification. For $\phi=\pi/2$, parallel states are connected by the probe pulse. The interaction is resonant and significant population transfer to excited states can occur. At the end of the strong pulse, whatever is the inversion of population reached at that time, the energy stored in the excited state is restored to the exciting field at longer time by FID. This explains the time dependence of the probe intensity envelope that exhibits a long ringing tail with decreasing amplitude as FID destroy the population in the excited states.

We also represent in Fig. 4, the results of numerical simulations showing explicitly the phase dependence of the probe intensity for the cases where $\phi=0,\pi/2$. These results confirm the experimental behaviour and the discussion above. The transmitted signal alternates between amplification and transparency regimes as the phase varies from $\phi=\pi/2$ to $\phi=0$.

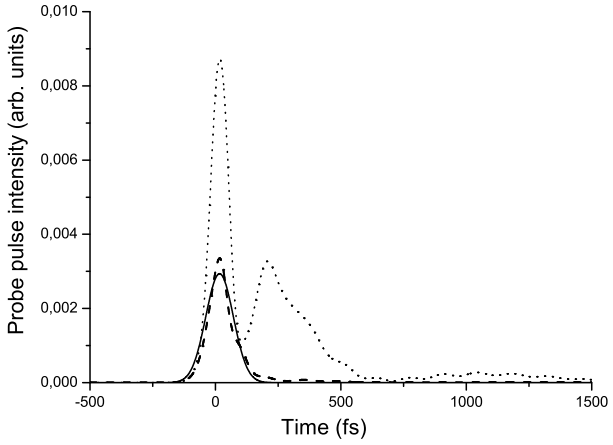

Figure 4: Theoretical curves obtained by numerical simulations for a set of parameters close to the experimental conditions. We have θ_1=2.2π, θ_2=0.1π, e_{disp}=0.2, and τ_0=85fs. An additional delay between the pulses of 30 fs and a chirp parameter about -3000fs^2 were taken into account. In dashed and dotted lines are plotted the curves obtained for $\phi=0$ and $\phi=\pi/2$ respectively whereas the curve in solid line represent the initial probe pulse.

5 Conclusion

In conclusion, we have studied the temporal profile of a weak σ-polarized pulse that propagates in a duplicated two-level system strongly driven by a π-polarized pulse. For $\phi = 0$, the interaction is non resonant in the adiabatic picture. The medium behaves as transparent and the probe pulse is almost unaffected as a consequence. For $\phi = \pi/2$, the interaction is resonant and the medium turns to be an amplifier. The excited state population radiates when returning back to the ground level and a long ringing tail is emitted that was experimentally observed and theoretically interpreted. Dispersion effects are generally considered as harmful because they distort the shape of the output pulse with few possibility of control [11–16].These results clearly show that the temporal profile of a light pulse can be manipulated by propagation effects thanks to a versatile experimental parameter ął the phase and a strong field regime [17, 18]. All these situations can be viewed as pump-probe experiments where the weak field probes the response of the system modified by the strong pulse. Electromagnetically induced transparency [19, 20] is one of the spectacular examples where the modification of the response of the system leads to a different propagation for the probe pulse, characterized by the formation of a transparency window and an anomalously strong deceleration [21].

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