

Coherent control of light shifts in an atomic medium driven by two orthogonally polarized pulses: Effect of the pulse overlap

J. C. Delagnes and M. A. Bouchene

Laboratoire de Collisions Agrégats Réactivité, C.N.R.S. UMR 5589, IRSAMC, Université Paul Sabatier, 118 Route de Narbonne, 31062 Toulouse Cedex 4, France

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We consider a duplicated two-level system where each two-level subsystem is driven by a strong resonant femtosecond pulse and a weak resonant femtosecond pulse connects cross transitions. Strong interference effects are induced in such a configuration leading to a modulation of the medium gain as a function of the relative phase ϕ between the pulses [J. C. Delagnes and M. A. Bouchene, Phys. Rev. Lett. **98**, 053602 (2007)]. We study here the influence of the temporal overlap between the pulses on the dynamics of the system and the medium gain. We show that the dynamics is dominated by two competing phenomena: Light shifts (LS) induced by the driving pulse that prevails for small delays and free induction decay (FID) that prevails for large delays. LS enhance the medium gain for $\phi=\pi/2$ and reduce it for $\phi=0$, whereas FID always leads to a decrease of the medium gain.

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I. INTRODUCTION

A lot of proposed methods to control physical and chemical processes rely on the interference effects produced by excitation with coherent fields. The control is performed by adjusting the relative phase between the exciting fields which steers the interference of the quantum paths involved in the process. Such control has been demonstrated in many contexts [1]. An interesting scenario is the possibility to control physical processes or pulse wave form when propagation effects are taken into account. Many previous studies have shown the dramatic modifications that occur in the behavior of the propagating pulses when the relative phase is modified [2–8]. If strong pulses are used, light shifts (LS) are induced in the medium and strong modification occurs in the atomic structure. LS are the basis for many spectacular phenomena in atomic and molecular physics. Rapid adiabatic passage, stimulated Raman adiabatic passage, Stark-chirped adiabatic passage [9] and light induced potential [10] are nonexhaustive examples of effects where LS play a crucial role. The possibility to control these effects has been demonstrated in only few cases. One can use shaped intense femtosecond pulses to realize a selective population of dressed states [11]. In a recent paper, we have demonstrated coherent control of LS effects in a duplicated two-level system [2]. A sequence of two femtosecond coherent pulses— π polarized *strong* pulse and σ polarized *weak* pulse—excite resonantly the $S_{1/2}-P_{1/2}$ transition of atomic rubidium in an optically dense sample. The σ pulse induces transitions between the adiabatic states with a coupling that is different for either identically or oppositely light-shifted states, and that can be modified by tuning the relative phase ϕ between the pulses. An efficient control of the medium gain for the σ pulse was experimentally demonstrated. It was shown to be the result of the interference between the absorption and the emission quantum paths for σ photons. In that experiment, the delay between the two pulses was smaller than the pulse durations so the pulse overlap was almost perfect. In this paper, we study the influence of the pulse overlap on the medium gain

and the LS effects. Spectacular changes occur with respect to the previous situation of perfect overlap. Indeed, the modification of the driving pulse area due to FID can no longer be neglected and LS effects are reduced. The competition between FID and LS leads to a significant modification of the medium gain. The situation $\phi=0$ for which destructive interference occurs at zero delay may be more favorable for amplification than the situation $\phi=\pi/2$ that was the previous favorable case.

II. COHERENT CONTROL OF LIGHT SHIFTS

We consider the situation of a duplicated two-level system $\{|1\rangle, |1'\rangle, |2\rangle, |2'\rangle\}$ [Fig. 1(a)] 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

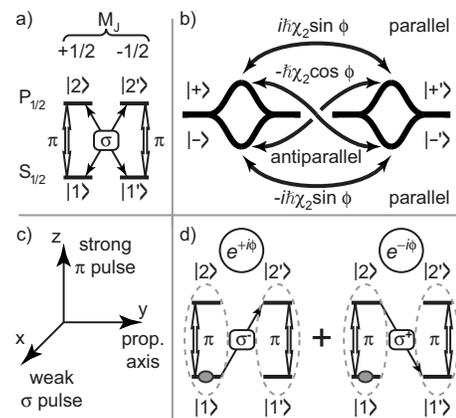


FIG. 1. (a) Energy levels and optical transitions involved in our system. (b) Adiabatic representation. (c) Polarization configuration. (d) Absorption and emission paths.

electric fields of the pulses that propagate along the y axis are expressed as $\vec{E}_\pi(y,t)=\vec{e}_z[\epsilon_{01}f_1(y,t)e^{-i\omega t}+\text{c.c.}]$ and $\vec{E}_\sigma(y,t)=\vec{e}_x[\epsilon_{02}f_2(y,t)e^{i\phi}e^{-i\omega t}+\text{c.c.}]$ with and $f_1(y=0,t)=\pi^{-1/2}\exp[-(t/\tau_0)^2]$, $f_2(y=0,t)=f_1(y=0,t-\tau)$, $\phi=\omega\tau$ is the relative phase between the pulses at the entrance of the medium, t represents the local time ($t=t_{\text{lab}}-y/c$), 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 $|1\rangle$.

Strong interference effects occur in the atomic system when the phase shift ϕ is varied in spite the fact that the two pulses are orthogonally polarized. The authors have already demonstrated in [2] that these interferences result from the superposition of two quantum paths, involving photon absorption from σ pulse on the transition $|1\rangle\rightarrow|2'\rangle$ and stimulated photon emission on the $|2\rangle\rightarrow|1'\rangle$ transition [Fig. 1(d)]. The interferences are observable only because the strong π -polarized pulse mixes both the initial states $\{|1\rangle,|2\rangle\}$ and the final states $\{|1'\rangle,|2'\rangle\}$ at the same time. This is a particularly original situation where interference effects take place between the two superposition states ($\{|1\rangle,|2\rangle\}\rightarrow\{|1'\rangle,|2'\rangle\}$). In the adiabatic representation [Fig. 1(b)], the interference effect is the result of the ϕ dependence of the coupling between the adiabatic states. At the entrance of the medium, the adiabatic states defined as $|\pm\rangle=(e^{-i\omega t}|2\rangle\mp|1\rangle)/\sqrt{2}$; $|\pm'\rangle=(-e^{-i\omega t}|2'\rangle\mp|1'\rangle)/\sqrt{2}$ are connected within the rotating wave approximation through the coupling elements:

$$\langle +|H_\sigma|+\rangle = -\langle -|H_\sigma|-\rangle = i\hbar\chi_2(t)\sin\phi, \quad (1)$$

$$\langle +|H_\sigma|-\rangle = -\langle -|H_\sigma|+\rangle = -\hbar\chi_2(t)\cos\phi, \quad (2)$$

with $\chi_2=d\epsilon_{02}f_2(0,t)/\hbar$, $d=\langle 1|\vec{d}\cdot\vec{e}_x|2'\rangle$ (\vec{d} is the dipole moment), and $H_\sigma=-\vec{d}\cdot\vec{E}_\sigma$. The parallel states with identical LS are thus coupled through the imaginary part of the weak field envelope (which is proportional to $\sin\phi$), whereas the antiparallel states with opposite LS are coupled through the real part (which is proportional to $\cos\phi$). The phase variation allows the modification of the coupling strength and the relative contribution between the transitions involving the parallel or antiparallel states performing a coherent control of LS. We now investigate how these interference effects modify the dynamics of the atomic system and affect the pulse behavior when these propagate through an atomic sample, in the optically dense regime.

A. Equation of propagation

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 linewidth. The absorption from the medium is thus negligible. Moreover, for long times, all the excited atoms return back to the ground state emitting coher-

ent radiation at a time scale much shorter than the excited states lifetime and the Doppler dephasing time (free induction decay). 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 [2,12]:

$$\frac{\partial(f_2(Y,t)e^{i\phi})}{\partial Y} = -i\frac{e_{\text{disp}}}{\theta_2}\rho^{(\sigma)}(Y,t), \quad (3)$$

with $Y=y/L$, L is the length of the medium, $\theta_2=(d\epsilon_{02}/\hbar)\tau_0$ is the pulse area of the σ pulse at the entrance of the medium (with $\theta_2\ll 1$), $e_{\text{disp}}=Nd^2\omega L\tau_0/2c\epsilon_0\hbar$ is a parameter that characterizes the severity of propagation effects, N is the atomic density, $\rho^{(\sigma)}=\rho_{2'1}+\rho_{21'}$ where $\rho_{ij}=\langle i|\rho|j\rangle$, $\rho=|\tilde{\psi}\rangle\langle\tilde{\psi}|$, $|\tilde{\psi}\rangle=e^{i(H_0/\hbar)t}|\psi\rangle$, H_0 is the Hamiltonian of the free atom, and $|\psi\rangle$ is the wave function. The coherence $\rho^{(\sigma)}$ can be expressed in the adiabatic representation as $\rho^{(\sigma)}=\rho_\pm\sin\phi+\rho_\mp\cos\phi$ with [2]

$$\rho_\pm(Y,t) = \cos\theta(Y,t)\int_{-\infty}^t\chi_2(Y,t')dt', \quad (4a)$$

$$\rho_\mp(Y,t) = -i\int_{-\infty}^t\cos\theta(Y,t')\chi_2(Y,t')dt', \quad (4b)$$

where $\theta(Y,t)=(-d\epsilon_{01}/\hbar)\int_{-\infty}^t f_1(Y,t')dt'$ is the truncated pulse area of the driving pulse at time t [the pulse area is $\theta_1(Y)=\theta(Y,+\infty)$, the envelope f_1 is real [13], and we denote in the following $\theta_1(0)$ θ_1]. With the standard notations, $-\cos\theta(Y,t)$ represents the inversion of population induced by the driving pulse between the ground and the excited states $|1\rangle$ and $|2\rangle$, respectively. Expressions (4a) and (4b) represent the contribution to the coherence of transitions between parallel and antiparallel states, respectively. The total transmitted energy is $W_{\text{out}}\propto\int_{-\infty}^{+\infty}I_2(Y=1,t)dt$. In the adiabatic picture, Eq. (4a) results from the fact that the coherence for the parallel states involves the superposition of two transition paths with both up-shifted and down-shifted states, respectively [Fig. 1(b)]. The transition amplitude of these two paths differs only by the free evolution phase $e^{-i\theta/2}$ and $e^{i\theta/2}$, respectively. The coherent superposition of the transition amplitudes leads to the cosine term in the expression of the radiating coherence ρ_\pm . In this situation, the coherence follows adiabatically the inversion of population created by the driving pulse. The contribution (4b) involves the overlap between the population inversion induced by the driving pulse and the σ pulse envelope. Thus the situation $\phi=0$ is analogous to that of a *single* two-level system where an inversion of population is created by a driving pulse and that is probed by a weak resonant pulse.

We assume for simplicity that during propagation, the π -polarized pulse is assumed to be weakly affected by the energy exchange with the σ pulse (numerical simulations account for exact depletion of the driving pulse). The dispersive effects slightly distort its shape because in the limit where $\theta_1\gg e_{\text{disp}}$ the number of incident photons exceeds largely the number of photons that interact with the atomic sample. Thus, the π pulse is transmitted with only small modifications of both its energy and shape. However, the

pulse area $\theta_1(Y)$ is significantly modified by propagation since the atom radiates as long as relaxations have not occurred. The radiated field is small at each instant but the time integral is no longer negligible. The pulse area obeys the McCall-Hahn theorem [13] derived from the equation of propagation of the driving pulse. It states that the pulse area evolves to an even multiple of π ($\theta_1(Y) \rightarrow 2n\pi$) as soon as $Y \gg \alpha_0^{-1}$ ($\alpha_0 = e_{\text{disp}}/\Delta_d \tau_0$ is the absorption coefficient at the line-center normalized to the length L of the medium and Δ_d is the Doppler width). In an optically dense medium ($\alpha_0 \gg 1$) and for long times, the atom returns back by FID to its ground level after propagation over a distance scale of the order of $Y \approx \alpha_0^{-1} \ll 1$ whatever is the initial strength of the driving pulse. This effect is important to understand the behavior of the atomic dynamics when the time delay is varied.

B. Case of perfect overlap

When the time delay is small enough such that the envelope of the two pulses can be considered coincident but with the phase-shift ϕ adjustable, interference effect with 2ϕ phase takes place [2]. For a fixed incident energy of the π -polarized pulse, the transmitted energy of the σ pulse oscillates with ϕ . It is maximum when $\phi = \pi/2$ and minimum for $\phi = 0$ in accordance with the respective constructive and destructive interference between the absorption and stimulated emission paths. In the situation where $\phi = \pi/2$, the medium gain [see relation (4a)] alternates between situations where amplification [$\cos \theta(Y, t) < 0$] or absorption [$\cos \theta(Y, t) > 0$] dominates the dynamics during the action of the strong field. At the end of the process, the σ pulse experiences generally a net positive gain except in the case where $\theta_1 < \pi/2$ for which the driving pulse draws more energy from the σ pulse than this latter can recover from FID at the end of the driving pulse. In the situation where $\phi = 0$, the contribution (4b) vanishes as the strength of the driving pulse increases such $\theta_1 \gg 2\pi$. In the adiabatic picture, the interaction of the σ pulse with the antiparallel states becomes gradually nonresonant as the LS increases. This leads to the appearance of a transparency window for the σ pulse that is almost unaffected by propagation.

An important remark can be done at this level. The σ pulse can lose its energy only by exchange with the strong pulse. 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 effect is illustrated in Fig. 2, where we represent the population dynamics of the atomic system for the set of parameters $\theta_1 = 1.5\pi$, $\theta_2 = 0.01\pi$, $e_{\text{disp}} = 0.18$, $\tau_0 = 90$ fs, $\tau = 0$, and $\phi = \pi/2$. These curves are obtained by solving numerically the Maxwell-Bloch equations for the atomic system (see the Appendix). The populations in the excited states $|2\rangle$ and $|2'\rangle$ reach nonvanishing asymptotic values when the exciting pulses ended. However, at the exit of the medium the populations exhibit different behavior. They decrease slowly in time exhibiting the usual ringing oscillations characteristics of FID behavior in an optically dense medium [13]. For long time no residual population subsists in these states. The behavior of the temporal shape

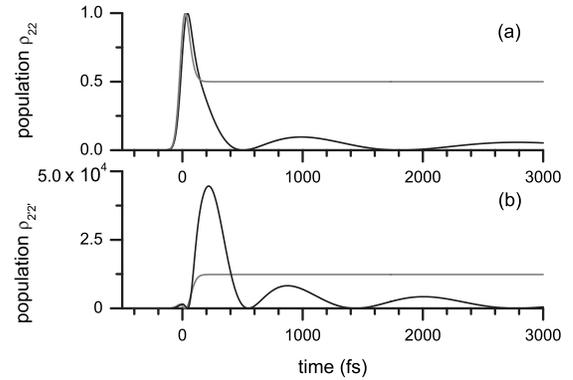


FIG. 2. Population dynamics ρ_{22} (a) and $\rho_{2'2'}$ (b) in excited states $|2\rangle$ and $|2'\rangle$, respectively. Curves are obtained by numerical simulations for the set of parameters $\theta_1 = 1.5\pi$, $\theta_2 = 0.01\pi$, $e_{\text{disp}} = 0.18$, $\tau_0 = 90$ fs, $\tau = 0$, and $\phi = \pi/2$. In gray and black lines are plotted the populations obtained at the entrance and the exit of the sample, respectively.

of the laser pulses shown in Fig. 3 is also instructive. The driving pulse is slightly distorted because of its sufficiently high strength whereas the σ pulse is amplified but exhibits a long ringing tail that decreases in time as FID destroy the population in the excited states.

C. Effect of temporal overlap

When the time delay is important, the partial overlap of the two pulses leads to drastic changes that can be understood from the behavior of the contributions in Eqs. (4a) and (4b) to the radiated energy. Let us distinguish again the two extreme situations $\phi = \pi/2$ and $\phi = 0$.

For $\phi = \pi/2$, only ρ_{\pm} can contribute to the transmitted energy. We show in a black solid line in Figs. 4(a) and 4(b) the variation of the transmitted σ pulse energy as a function of time delay for two energies of the driving pulse such as $\theta_1 = 0.75\pi$ and 1.5π , respectively. In both cases, the transmitted energy slowly decreases with the time delay as the pulse overlap vanishes. The asymptotic value on both sides corresponds to the input energy of the σ pulse. This can be un-

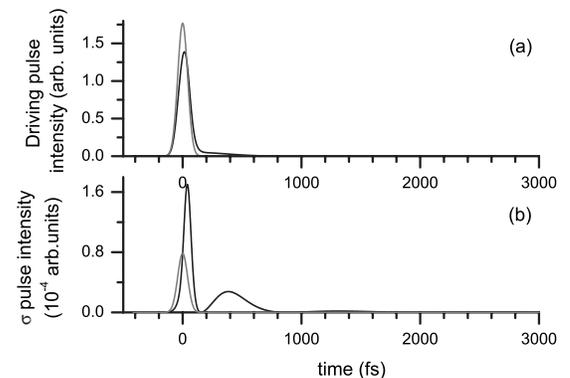


FIG. 3. Temporal behavior of the (a) driving pulse and (b) σ pulse. The parameters are those given in Fig. 2. In gray and black lines are plotted the intensity obtained at the entrance and the exit of the sample, respectively.

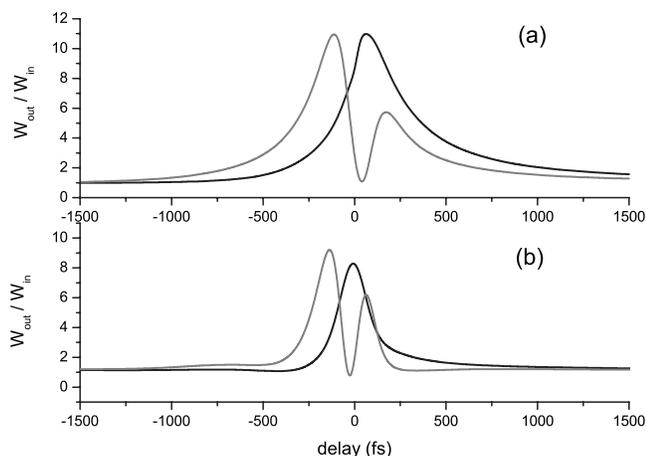


FIG. 4. Normalized transmitted σ pulse energy with $\theta_2=0.01\pi$, $e_{disp}=0.18$, and $\tau_0=90$ fs for (a) $\theta_1=0.75\pi$ and (b) $\theta_1=1.5\pi$. In gray and black lines are plotted the curves obtained for $\phi=0$ and $\phi=\pi/2$, respectively.

derstood as follows. For $\tau \rightarrow -\infty$, the σ pulse excites the system before the action of the driving pulse. Although the medium is an absorber the total absorption is small since the relaxations are negligible. All the energy stored in the excited states by absorption of the σ pulse photons is restored back coherently to this field by FID. The situation where $\tau \rightarrow \infty$ leads to *a priori* paradoxical situation for $\theta=0.75\pi$: Although the driving pulse induces a significant inversion of population at the entrance of the medium and one therefore expects an amplification of the σ pulse, this latter is transmitted without any modification of its energy. This is again the consequence of the application of the pulse area theorem to the driving pulse: The pulse area of this latter reaches $2n\pi$ as soon as $Y \gg \alpha_0^{-1}$ and no inversion of population can persist for long times in the system as a consequence. The situation where the pulses partially overlap represents an intermediate case where the inversion of population is partly destroyed by FID reducing the exchange of energy between the fields.

For $\phi=0$, only ρ_{\neq} contributes to the transmitted energy. We show in gray solid line in Figs. 4(a) and 4(b), the behavior of the transmitted energy when the time delay between the two pulse is varied with $\theta_1=0.75\pi$ and 1.5π , respectively. In both cases, the signal initially at minimum around zero delay increases with the modulus of the delay. It reaches a maximum and then decreases again to an asymptotic value equal to one because of FID as explained in the previous case ($\phi=\pi/2$). The minimum value in Fig. 4(b) is obtained for $\tau \approx -30$ fs where the transmitted energy is less than the input energy (the ratio is 0.55). Here, the exchange of energy between the pulses is at the expense of the σ pulse that loses its energy with the gain of the driving pulse. The original situation for $\phi=0$ is the region of intermediate delays, where two maximums appear. Although the interaction with the σ pulse is nonresonant (in the adiabatic picture), a significant gain is obtained. The signal may even be higher than that obtained for $\phi=\pi/2$ where the interaction is resonant. Indeed, the partial overlap results in the reduction of the LS effects. Amplification of the σ pulse is then possible and a maximum can be obtained by adjusting the delay. At higher

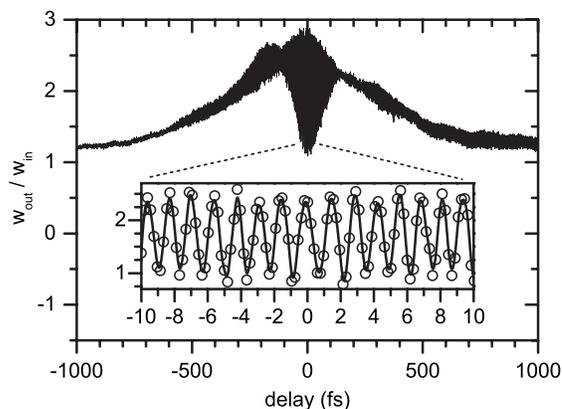


FIG. 5. Experimental results. Dependence of the transmitted σ pulse energy with the delay for $\theta_1 \approx 1.5\pi$. Inset exhibits the rapid variation of the signal (1.35 fs period) due to interferences between absorption and stimulated emission paths. See text for parameter values.

delay, the FID leads to a vanishing inversion of population and the transmitted energy remains unaffected as explained above.

D. Experiment and results

Experimental observation of these effects has been achieved in rubidium on the $S_{1/2}-P_{1/2}$ transition (794.7 nm). A regenerative amplifier pumped by a titanium sapphire laser delivers linearly polarized laser pulses with 100 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 and the energies are about $60 \mu\text{J}$ and $0.14 \mu\text{J}$, respectively. This leads to pulse areas $\theta_1=1.5\pi$ and $\theta_2=0.1\pi$. The heat pipe (length 12 cm) was heated at a temperature $T=135^\circ\text{C}$ for which $e_{disp}=0.18$. The dimensionless absorption coefficient (optical depth) is $\alpha_0=7500 \gg 1$. At the exit, the two pulses are separated by a polarizer and the energy of the weak transmitted pulse is measured with a photodiode. Experimental results are displayed in Fig. 5. The energy of the σ pulse is modulated with a period of about 1.36 fs, close to half of the optical period (2.65 fs) consequence of the 2ϕ interference. Because our interferometer is not stabilized, we were not able to fix the value of the relative phase for sufficient long time in order to acquire the complete curve giving the dependence of the transmitted σ pulse energy with the delay. The envelopes of the experimental curves in Fig. 5 are related to the theoretical curves in Fig. 6. These latter are obtained taking into account for spin-orbit effects due to the presence of the $P_{3/2}$ state and that can modify the gain value [14]. Good agreement is obtained.

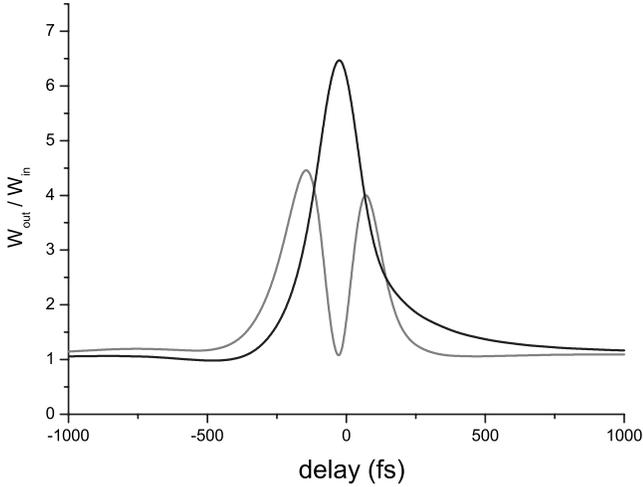


FIG. 6. Theoretical curves obtained by numerical simulations for a set of parameters close to the experimental conditions (Fig. 5). We have $\theta_2=0.1\pi$, $e_{\text{disp}}=0.18$, $\tau_0=90$ fs, and $\theta_1=1.5\pi$. In gray and black lines are plotted the curves obtained for $\phi=0$ and $\phi=\pi/2$, respectively.

Moreover, the nodes around $\tau \approx -150$ fs and $\tau \approx 100$ fs where the $\phi=\pi/2$ curve crosses the $\phi=0$ curve becoming lower in 4 are exhibited in the experimental curve even if not completely resolved. The maximum gain obtained for $\tau \approx 0$ and $\phi=\pi/2$ is lower than that predicted theoretically. This is mainly due to a nonperfect spatial overlap and a residual chirp of the pulses that both modify the effective phase shift and the gain.

III. CONCLUSION

In conclusion, we have studied the exchange of energy between two orthogonally polarized pulses (strong driving π -polarized pulse and weak propagating σ -polarized pulse) in a duplicated two-level system. We study the transmitted energy of the σ pulse as a function of the overlap between the pulses, the relative phase, and the driving pulse energy. Significant differences occur when comparing the results with the standard case of a perfect overlap studied previously

[2]. In this latter situation, the transmitted signal alternates between amplification and transparency regimes as the phase varies from $\phi=\pi/2$ to $\phi=0$, switching from a resonant to a nonresonant interaction of the σ pulse in the adiabatic picture. Here, two noticeable effects are demonstrated. First, the asymptotic value of the output energy obtained for large delay τ ($|\tau| \rightarrow \infty$) is equal to the input energy even if the driving pulse causes a total inversion of population at the entrance of the medium. This is shown to be a direct consequence of the McCall-Hahn theorem that states that the pulse area of the driving pulse goes to an even number of 2π in a very short penetration distance. The excited state population returns back to the ground level by FID making the stimulated emission from excited states vanishes. The second effect is observed for $\phi=0$. Although the interaction of the σ pulse is non resonant (in the adiabatic representation) significant amplification is obtained for this pulse when the overlap with the driving pulse is not perfect reducing the LS effects. The gain obtained can be even higher than the one obtained for $\phi=\pi/2$ where the interaction is resonant in the adiabatic representation.

APPENDIX

Theoretical curves are obtained by solving by the Maxwell-Bloch equations for the atomic system coupled to the laser fields (including the driving pulse). Moreover, the σ pulse obeys to Eq. (3) and the driving pulse to the equation:

$$\frac{\partial f_1(Y,t)}{\partial Y} = i \frac{e_{\text{disp}}}{\theta_1} \rho^{(\pi)}(Y,t), \quad (\text{A1})$$

with $\rho^{(\pi)} = \rho_{21} - \rho_{2'1'}$, where $\rho_{ij} = \langle i | \rho | j \rangle$, $\rho = |\tilde{\psi}\rangle\langle\tilde{\psi}|$, $|\tilde{\psi}\rangle = e^{i(H_0/\hbar)t} |\psi\rangle$, H_0 is the Hamiltonian of the free atom, and $|\psi\rangle$ is the wave function. The evolution of the density matrix obeys to the following equation:

$$\partial_T \rho = - \frac{i}{\hbar} [H, \rho], \quad (\text{A2})$$

with $\partial_T = \partial/\partial T$, $T = t/\tau_0$, and H expresses in the basis set $\{|1\rangle, |1'\rangle, |2\rangle, |2'\rangle\}$ as

$$H = -\hbar \begin{pmatrix} 0 & 0 & \theta_1 f_1^*(Y,T) & \theta_2 f_2^*(Y,T) e^{-i\phi} \\ 0 & 0 & \theta_2 f_2^*(Y,T) e^{-i\phi} & -\theta_1 f_1^*(Y,T) \\ \theta_1 f_1(Y,T) & \theta_2 f_2(Y,T) e^{i\phi} & 0 & 0 \\ \theta_2 f_2(Y,T) e^{i\phi} & -\theta_1 f_1(Y,T) & 0 & 0 \end{pmatrix}. \quad (\text{A3})$$

The set of Eqs. (3), (A1), and (A2) is solved numerically using partial differential equations solver (D03PEF routine of NAG fortran library) with the initial conditions $f_1(Y, T \rightarrow -\infty) = f_2(Y, T \rightarrow -\infty) = 0$ and $\rho_{ij}(Y, T \rightarrow -\infty) = \delta_{i1} \delta_{j1}$. The boundary conditions are $f_1(Y=0, T) = f_2(Y=0, T+\tau) = \pi^{-1/2} \exp(-T^2)$ for the fields whereas for the atomic quantities the boundary conditions are obtained by solving Eq. (A2) at the entrance of the medium using a fourth-order Runge-Kutta method.

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