Research Signpost
37/661 (2), Fort P.O., Trivandrum-695 023, Kerala, India

Aspects of Optical Sciences and Quantum Information, 2007: 173-193
ISBN: 81-308-0147-7 Editor: Mahmoud Abdel-Aty

# 6 Application of propagation effects in atomic vapours for the shaping of ultrashort pulses

**J. C. Delagnes, F. A. Hashmi and M. A. Bouchene**
Laboratoire de Collisions Agrégats Réactivité, CNRS, UMR 5589,IRSAMC
Université Paul Sabatier, 118 Route de Narbonne, 31062 Toulouse Cedex 4
France

## Abstract

*The studies of coherent control experienced a considerable development during this last decade. The successful progress in the technology of « pulse-shapers » which modify on request the spectrum of the laser pulses, allowed the control of various physical and chemical processes. However, these devices do not remain useful in picosecond and VUV domains because of the narrower spectrum width of the former and high frequency range of the latter. Additionally, these devices are of passive type and cannot introduce new frequency components in the spectrum.*

Correspondence/Reprint request: Dr. M. A. Bouchene, Laboratoire de Collisions Agrégats Réactivité, CNRS UMR 5589,IRSAMC, Université Paul Sabatier, 118 Route de Narbonne, 31062 Toulouse Cedex 4, France
E-mail: aziz@irsamc.ups-tlse.fr

*We review in this paper an alternative method using the re-shaping phenomena experienced by an ultrashort pulse as it propagates through an optically dense atomic medium. We specially focus on the situation of atoms driven by an additional strong pulse that modify drastically and in a controllable way the optical response of the medium allowing the re-shaping of the propagating pulse.*

# I. Introduction

Pulse-shaping methods allow the synthesis of complex femtosecond pulses with arbitrary spectral waveforms according to user specifications [1]. The technical improvement of these methods has boosted many fields of research in physics and chemistry. For instance, in femtosecond optics compression of phase-modulated femtosecond pulses has been achieved [2] and tailored femtosecond pulses has been used to optimize high-harmonic generation in gases [3]. In quantum control domain, these tailoring methods combined with optimisation techniques have allowed optimal control of molecular and chemical processes. One challenge of these studies is to synthesize chemical substances with high efficiency while at the same time reducing unwanted by-products. The process works as follows. A laser pulse passes through a pulse-shaper, it is then shone on a sample (atoms or molecules) and products like electrons or molecular fragments are obtained from the physical or chemical processes induced by the laser (dissociation, ionization, or induced chemical reactions). The aim of the control is to optimise a desired output (generally the signal of a desired fragment) by modifying the applied laser field. A suitable algorithm that pilots the pulse-shaper forms an ultrashort pulse, uses the desired output as feedback in a learning algorithm, and iteratively improves the applied laser field until the convergence of the procedure. Experimental realization of the automated optimization has been achieved in 1998 in the case of photofragmentation of $CpFe(CO)_2Cl$ by Gerber *el al* [4]. They showed that two different bond-cleaving reactions can be selected, resulting in chemical different products.

Several pulse-shaper architectures have been proposed. Most all of them rely on the spatial dispersion of the frequency spectrum of an ultrashort laser pulse. The shaping, which consists in the modification of the amplitude and/or phase of these components is achieved thanks to devices such as liquid crystal displays (LCD) [5] acousto-optics modulator (AOM) [6], and deformable mirrors [7]. After being suitably tailored, the spectral components are recombined spatially at the exit of the device. Although these devices have shown a great efficiency in the tailoring of ultrashort pulses, they suffer from several drawbacks. For instance, LCD and AOM don’t operate in the VUV domain where they are opaque, and their spectral resolution limits generally the shaping to very broadband pulses (femtosecond pulses). They are thus inadequate to shape relatively long pulses. We review in this paper an alternative method using the re-shaping phenomena experienced by an ultrashort pulse as it propagates through an optically dense atomic medium. We specially focus on the situation of atoms driven by an additional strong pulse that modify dramatically the optical response of the medium allowing the re-shaping of the propagating pulse [8, 9]. The paper is structured as follows. In chapter II, we recall the basics of the semi-classical interaction of a strong laser pulse with a two-level atom by introducing the adiabatic description. We recall also the basics of the propagation of a weak ultrashort pulse in an optically dense medium. In chapter III, we study the shaping effects obtained by the combination of dispersion effects and light-shifts. We consider in III-1, the situation where a sequence of two pulses –a non resonant pulse with strong intensity and a resonant pulse with weak intensity- excite an assembly of two-level atoms (fig.1-a). In part III-2, the same laser sequence is considered but the weak pulse propagates resonantly on the $|0\rangle \rightarrow |1\rangle$ transition of a three-level system $(|0\rangle,|1\rangle,|2\rangle)$ while the strong field is applied non-resonantly on the $|1\rangle \rightarrow |2\rangle$ transition (fig.2-a). In both cases, important light-shifts are induced. We theoretically study the behaviour of the weak propagating laser pulse in such conditions. For the transmitted field profile, drastic changes occur: at long time scale, efficient reduction of the distortion is obtained, while strong oscillations appear for short times mapping out the dynamical light-shift. These shaping effects accompanied with new features in the spectrum for the propagating pulse are interpreted as the result of the interference between the incident field and the induced dipole radiated field, whose frequency sweeps in time because of the induced light-shift. In chapter IV, an analogous effect is described with the propagation of a chirped pulse with weak intensity in a two-level system [10]. Finally, a conclusion ends this paper.

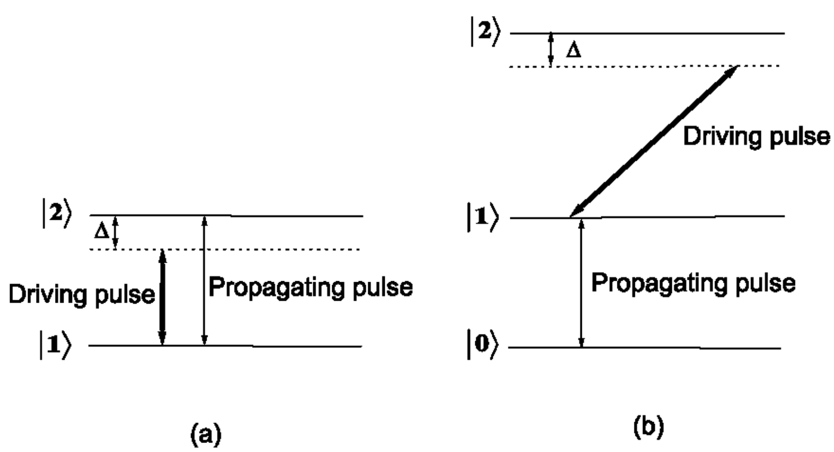


**Figure 1.** Schematic representation of the (a) two-level, (b) three-level system in the bare-state picture.

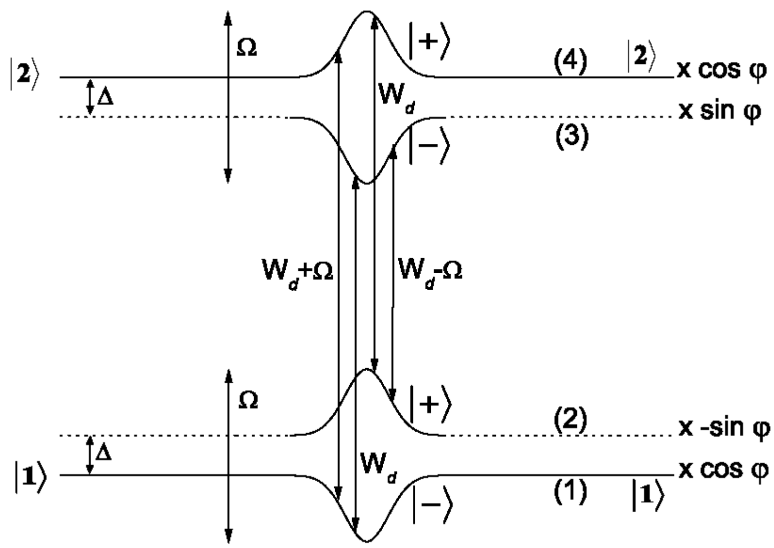


**Figure 2.** Adiabatic representation of the two-level system: each stationary state can be decomposed as a linear superposition of the two adiabatic states with time dependent energies. Three resonance frequencies appear at $W_d$ (solid arrows), $W_d + \Omega$ (dashed arrow), $W_d - \Omega$ (dotted arrow).

# II. Adiabatic states. Equation of propagation

## II.1 Adiabatic states

We consider a two-level system (states $|1\rangle$ and $|2\rangle$) with energies 0 and $\hbar\omega_0$. The system is driven by a strong ultrashort pulse whose electric field is expressed as:

$$E_d(z,t) = \frac{1}{2}\varepsilon_{0d} f_d(t) e^{-i(\omega_d t - k_d z)} + c.c. \tag{1}$$

$\varepsilon_{0d}$ is the field amplitude and $f_d$ is the envelope of the pulse. We designate by c.c the complex conjugate. Throughout this paper, we consider that the envelope of the pulse is real and expresses as $f_d(t) = \frac{1}{\sqrt{\pi}} e^{-\left(t/\tau_d\right)^2}$ where $\tau_d$ represents the temporal width of the pulse at the entrance and $\tau_d^{-1}$ is the spectral bandwidth of the pulse. The central frequency of the pulse $\omega_d$ is detuned from the resonance $\omega_0$ and $\omega_d = k_d c$. We introduce dimensionless time and space variables $Z$ and $T$ as $Z = z/L$ and $T = (t - z/c)/\tau_d$. Here, $L$ is the length of the sample through which the pulse propagates. In this first part, we assume that the strong pulse contains enough photons to avoid the effect of the radiated field on it. No modification of the pulse envelope is thus expected. Exact condition for such behavior is established in §II-2. Dimensionless forms of the frequencies are $W_d = \omega_d \tau_d, W_o = \omega_o \tau_d$. The wavefunction of the system can be written as :

$$|\psi\rangle(T) = a_1(T)|1\rangle + a_2(T)e^{-iW_dT}|2\rangle \tag{2}$$

For an ultrashort pulse, the relaxation and the Doppler dephasing effects can be neglected because of their long time scale. Using the Schrödinger equation and within the rotating wave approximation (RWA), we obtain the following equations for the evolution of atomic quantities ($\partial_T \equiv \frac{\partial}{\partial_T}$)

$$i\partial_T \begin{pmatrix} a_1 \\ a_2 \end{pmatrix}(T) = \begin{pmatrix} 0 & -\theta_d f_d^*/2 \\ -\theta_d f_d/2 & \Delta \end{pmatrix} \begin{pmatrix} a_1 \\ a_2 \end{pmatrix}(T) \tag{3}$$

Here $\Delta = (\omega_0 - \omega_d)\tau_d$ is the dimensionless detuning and $\theta_d = \mu\varepsilon_{0d}\tau_d/\hbar$ is the usual pulse area that characterizes the strength of the pulse.

An alternative description of the interaction in which the effect of the field in the non-resonant case is more clearly exhibited is possible with the use of the adiabatic basis (eigenbasis of fully perturbed Hamiltonian within the RWA). We recall here some of the important properties.

We define the new basis by the following transformation:

$$\begin{aligned} |-\rangle(T) &= \cos\phi(T)|1\rangle + \sin\phi(T)\, e^{-iW_dT}|2\rangle \\ |+\rangle(T) &= -\sin\phi(T)|1\rangle + \cos\phi(T)\, e^{-iW_dT}|2\rangle \end{aligned} \tag{4}$$

With the mixing angle $\phi$ given by

$$\tan(2\phi) = \frac{\theta_d}{\Delta} f_d(T) \tag{5}$$

In the new basis the wave function can be written as

$$|\psi\rangle(T) = \alpha_-(T)|-\rangle(T) + \alpha_+(T)|+\rangle(T) \tag{6}$$

The amplitudes in the two basis set are related by the following relations (equations 2, 4 and 6),

$$a_1(T) = -\alpha_+(T)\sin\phi(T) + \alpha_-(T)\cos\phi(T) \tag{7-a}$$

$$a_2(T) = \alpha_+(T)\cos\phi(T) + \alpha_-(T)\sin\phi(T) \tag{7-b}$$

The amplitudes $\alpha_+$ and $\alpha_-$ evolve according to the equation (equations 3 and 7)

$$i\partial_T \begin{pmatrix} \alpha_- \\ \alpha_+ \end{pmatrix} = A \begin{pmatrix} \alpha_- \\ \alpha_+ \end{pmatrix} \quad (8)$$

with $A = \begin{pmatrix} (-\Omega+\Delta)/2 & i\partial_T\phi \\ -i\partial_T\phi & (\Omega+\Delta)/2 \end{pmatrix}$.

Here, $\Omega$ is the dimensionless generalized Rabi frequency defined as $\Omega = \sqrt{\theta_d^2 f_d^2 + \Delta^2}$. The diagonal terms $\frac{1}{2}(\Delta \pm \Omega)$ represent the adiabatic energy levels and $\partial_T\phi$ is the non-adiabatic coupling which depends strongly on the shape of the pulse. $\Omega$ gives the instantaneous separation of the adiabatic levels becoming $\Delta$ as $T$ approaches $\pm\infty$. The adiabatic levels experience a transient light-shift during the action of the driving pulse.

In the resonant case $\Delta = 0$, we thus have $\Omega = \theta_d\ f_d, \phi = \pi/4$ and $\partial_T\phi = 0$. Initially, the population is equally distributed between the two adiabatic states i.e. $\alpha_\pm(T \to -\infty) = \mp 1/\sqrt{2}$ and at time T the amplitudes are given by the expression $\alpha_\pm(T) = \mp 1/\sqrt{2}\ e^{\mp i\frac{\theta_d}{2}\int_{-\infty}^{T} f_d dT'}$. The population in the states $|1\rangle$ and $|2\rangle$ which are linear superposition of the adiabatic states exhibits Rabi oscillations.

In the non-resonant case and when the off-diagonal term can be neglected, the evolution is said to be adiabatic. The system starts from stationary states at $T \to -\infty$, i.e. $|-\rangle(T \to -\infty) = |1\rangle$ and $|+\rangle(T \to -\infty) = e^{-iW_dT}|2\rangle$. The driving pulse introduces new states during the transient time and as $T \to \infty$, the system moves back to the original configuration. If initially all the population is in the ground state $|1\rangle$, during the pulse excitation some transient population appears in the state $|2\rangle$, but at the end of the pulse all population returns back to the ground level. The important feature of this evolution is that no net transfer of population to state $|+\rangle$ occurs. However, in the presence of a non-diagonal coupling term, non-adiabatic transitions to the adiabatic state $|+\rangle$ results in asymptotic population in state $|2\rangle$. The non-adiabaticity of the excitation process is usually characterized by evaluating the inverse Massey "parameter" defined as the ratio between the coupling term $\partial_T\phi$ and the generalized dimensionless Rabi frequency $\Omega$ [11]:

$$M^{-1} = \partial_T\phi/\Omega \quad (9)$$

For $M^{-1} \ll 1$, the evolution can be considered as adiabatic and the system experiences only light-shifts. Otherwise, the interaction leads to non-adiabatic population transfer that depends strongly on the shape of the pulse through $\partial_T \phi$ [11].

## II.2 Equation of propagation

Let's consider now an optically dense sample of two-level atoms through which an ultrashort resonant laser pulse with weak intensity propagates. The expression of this field is given by:

$$E_p\left(\vec{r},t\right)=\frac{1}{2}\varepsilon_{0p}\,f_p\left(z,t\right)e^{-i\left(\omega_p t-\vec{k}_p\vec{r}\right)}+c.c. \qquad (10)$$

$\varepsilon_{0p}$ is the field amplitude (real) and $f_p$ is the Gaussian envelope of the pulse with a pulse duration $\tau_p$. The pulse envelope may be distorted during propagation because of its weak intensity and of the resonant character of the interaction ($\omega_p=\omega_0$). The propagation of the pulse obeys the Maxwell equations. Neglecting the diffraction effects, the Doppler effect and within the slowly varying envelope approximation, the electric field evolves as [12]:

$$\frac{\partial}{\partial Z}f_p=i\frac{e_{disp}}{\theta_p}a_1^*a_2 \qquad (11)$$

The dimensionless coefficient $e_{disp}=\dfrac{n\mu^2\,\omega_o}{2c\,\varepsilon_0\,\hbar}\,L\,\tau_p$ characterizes the severity of propagation effects on the $|1\rangle-|2\rangle$ transition. It depends on the density of atoms $n$ and the oscillator strength through the square of the dipole moment $\mu^2$. Moreover, introducing the coefficient of absorption at resonance $\alpha_0=\dfrac{n\mu^2\,\omega_o}{2c\,\varepsilon_0\,\hbar\,\Delta_d}$ where $\Delta_d$ is the Doppler width, and the optical depth parameter $\alpha_0 L$, we have the relation $e_{disp}=\alpha_0\,L\,\Delta_d\,\tau_p$. The quantity $\alpha_0\,L\Delta_d$ represents the spectral domain around the resonance over which the dispersion affects the spectral phase of the incident pulse [13]. Thus, $e_{disp}=\alpha_0\,L\,\Delta_d\,/\,\tau_p^{-1}$ can be interpreted as the ratio between this spectral range and the spectral bandwidth of the pulse. The modification of the pulse through propagation is due to the radiating dipole arising from the coherence $a_1^*a_2$ as shown by equation (11). For a pulse far from resonance, the coherence is strongly reduced and the distortion of the pulse is small (unless the density $n$ is very high). For a resonant pulse in the weak field regime $\left(\theta_p<<1\right)$, we get $\left|a_1^*a_2\right|\le\theta_p/2$ and the distortion of the field becomes important when $e_{disp}\ge 1$.

This latter inequality means that dispersion alters the phase of all the spectral components of the incident pulse and so propagation effects can not be neglected. For a resonant pulse in the strong field regime ($\theta_p \geq 1$), the amplitude of the coherence can not exceed its maximum value ½ and the propagation effects are small when $\theta_p >> e_{disp}$ (even if $e_{disp} \geq 1$). Note that, the total absorption is always negligible for ultrashort pulses since their spectral bandwidth is much larger than the Doppler width [13] and the dominant effect is dispersion. The solution of equation (19) can be obtained formally [14] as:

$$f_p(t,1) = f_p(t,0) + f_{rad}(t,1) \tag{12}$$

with:

$$f_{rad}(t,1) = \int_{-\infty}^{+\infty} R(t')\, f_p(t-t',0)\ \ dt' \tag{13}$$

It represents the convolution between the incident field and a causal response function $R(t')$ ($R(t'<0)=0$) and expresses for $t'>0$ as :

$$R(t') \simeq -\frac{2e_{disp}}{\tau_p} \frac{J_1\left(2\sqrt{e_{disp}t'/\tau_p}\ \right)}{\left(2\sqrt{e_{disp}t'/\tau_p}\ \right)} \tag{14}$$

$J_1$ is the Bessel function of first order. In the time domain, the ultrashort pulse presents strong ringing oscillations with a time scale on the order of $\tau_p / e_{disp}$.

## III- Propagation in a driven atomic system of a resonant weak ultrashort pulse

We have shown in the preceding section that the driving pulse induces transient light-shifts in a two-level system. In this section we will show how these transient light-shifts can be used to modify the shape of another weak ultrashort pulse. We will present two cases in this respect. First we consider that the two-pulse sequence -the strong non resonant driving pulse and the weak resonant propagating pulse- interacts with the same two-level system (fig. 1-a). The two pulses cross at small angle, so that they overlap in the interaction region but remain spatially separable. In the second case, we consider a three-level ladder system in which the driving pulse induces light-shifts on two excited levels and the weak propagating pulse couples one the ground level to one of the excited states (fig. 1-b). In both cases, we assume that because of the high intensity of the driving pulse and the non-resonant character of the interaction, the atomic system experiences a purely adiabatic evolution and the driving pulse is prevented from any distortion due to propagation effects.

### III-1 Two-level system situation

We assume that at the entrance of the medium, the propagating resonant pulse is coherent with the non-resonant driving pulse but has a different direction of propagation. We assume also that the angle between the wave vectors is small enough to use the one dimensional approximation. We take the same pulse shape for both pulses but the two can have different pulse duration. The electric fields of the driving and propagating pulses express as in relations (1) and (10) respectively with the relation:

$$f_p(z=0,t) = f_d\left(z=0,t/\tau_{pd}\right)e^{i\beta} \tag{15}$$

where $\tau_{pd} = \tau_p/\tau_d$ is the ratio between the duration of the propagating and the driving pulse and $\beta$ is the phase shift between the two coherent pulses.

The Schrödinger equation in the adiabatic state representation now becomes:

$$i\partial_T\begin{pmatrix}\alpha_- \\ \alpha_+\end{pmatrix} = \left(A+V\right)\begin{pmatrix}\alpha_- \\ \alpha_+\end{pmatrix} \tag{16}$$

with $V = V^{(+)}e^{i\delta\vec{k}\vec{r}} + V^{(-)}e^{-i\delta\vec{k}\vec{r}}$ the perturbation matrix due to the weak field and $V^{(+)}$ is given by:

$$V^{(+)} = \frac{\theta_p}{2\tau_{pd}} f_p\, e^{-i\Delta T}\begin{pmatrix}-\dfrac{\sin 2\phi}{2} & \sin^2\phi \\ -\cos^2\phi & \dfrac{\sin 2\phi}{2}\end{pmatrix} \tag{17}$$

and $V^{(-)} = \left(V^{(+)}\right)^+$. Here, $\delta\vec{k} = \vec{k}_p - k_p\,\vec{e}_z$, and $\theta_p = \mu\,\varepsilon_{0p}\tau_p/\hbar$ represents the pulse area of the propagating pulse. In this representation, the perturbation matrix has both diagonal and non-diagonal terms that depend on both the driving and the propagating pulses. In the presence of the driving pulse the dipole moment felt by the propagating pulse is modified. The diagonal terms induce self phase modulation. These contributions are small when compared to the light-shift as long as the propagating field is weak ($\theta_p \ll 1$).

In the perturbative regime, the coherence can be developed up to the first order with respect to the propagating field amplitude as:

$$a_1^* a_2 \simeq \rho_d + \rho_p\, e^{i\delta\vec{k}\vec{r}} + \rho_p'\, e^{-i\delta\vec{k}\vec{r}} \tag{18}$$

$\rho_d$ , $\rho_p$ and $\rho_p'$ are responsible for the radiation in the direction of the driving pulse $\vec{k}_d$, propagating pulse $\vec{k}_d + \left(\delta\vec{k} + \frac{\Delta}{c\,\tau_d}\vec{e}_z\right)$ and the symmetric direction $\vec{k}_d - \left(\delta\vec{k} + \frac{\Delta}{c\,\tau_d}\vec{e}_z\right)$ respectively. The equation of propagation for the weak field can be written as:

$$\partial_Z \left( f_p\, e^{-i\Delta T} \right) = i\frac{e_{disp}}{\theta_p}\rho_p \tag{19}$$

Since the frequency of the propagating pulse is no longer resonant when the driving pulse is applied, the expression of the coherence $\rho_p$ can be easily derived. This simplification can be understood using the adiabatic representation (fig. 2). Indeed, we can associate eight different quantum paths contribute to the coherence. Four correspond to absorption of the propagating pulse from level 1 and 2 to 3 and 4, while the four others correspond to emission in the opposite direction. The associated transition frequencies are $W_d$ $(1\leftrightarrow 3, 2\leftrightarrow 4)$, $W_d - \Omega\,(2\leftrightarrow 3)$ and $W_d + \Omega$ $(1\leftrightarrow 4)$. The propagating pulse is no longer resonant except in a specific situation corresponding to paths $1\leftrightarrow 4$ and restricted to time T before and after the application of the driving pulse. This necessitates $\tau_{pd} > 1$. Now if all the population is initially in the ground state (level 1), 4 is not populated during adiabatic evolution and only the absorption path $(1\rightarrow 4)$ is efficiently involved. From relations (7, 16-18), the contribution to the coherence $\rho_p$ and corresponding to the absorption path $1\rightarrow 4$ is:

$$\rho_p \simeq -i\cos^2\phi \int_{-\infty}^{T} V_{+-}^{(+)}(T',Z)\, e^{-i\int_{T'}^{T}\Omega\, dT''}\, dT' \tag{20}$$

We note by -T$_0$ and T$_0$ the solutions of the equation $\Omega - \Delta = \tau_{pd}^{-1}$. This time interval [-T$_0$, T$_0$ ] represents the duration for which the light-shift between the adiabatic states induced by the driving pulse is sufficiently strong to make the propagating pulse non-resonant. Using relations (16, 17 and 20), the coherence $\rho_p$ for time T with $-T_0 \le T \le T_0$ can be approximated by the following term corresponding to the resonant contribution that builds up from $-\infty$ to $-T_0$:

$$\rho_p \simeq \frac{i\theta_p}{2\tau_{pd}}\cos^2\phi\, e^{-i\Delta T}\, e^{-i\int_{-\infty}^{T}(\Omega-\Delta)\, dT'} \int_{-\infty}^{-T_0} f_p(Z,T')dT' \tag{21}$$

The transmitted propagating field is obtained by adding the incident field to the radiated one. Using equations (19) and (21), the transmitted intensity $I_p(1,T)=\left|\varepsilon_{0p}\ f_p(1,T)\right|^2$ can be approximated for $-T_0 \le T \le T_0$ at the lowest order of the dispersion parameter by the following expression:

$$I_p(1,T)\simeq I_{0p}\left[\left|f_p(0,T)\right|^2 - e_{disp}\cos^2\phi(T)\ \left(\cos\left(\int_{-\infty}^{T}(\Omega-\Delta)\ dT'-\beta\right)\right)\left|\int_{-\infty}^{-T_0} f_p(0,T')d\left(\frac{T'}{\tau_{pd}}\right)\right|\right] \tag{22}$$

with $I_{0p}=\left|\varepsilon_{0p}\right|^2$. Formula (22) shows that the transmitted pulse intensity is modulated with an interference pattern (cosine term in bracket) which depends on the light-shift induced on the transition $1\rightarrow 4$ (fig. 2). These oscillations may be shifted by varying the relative phase-shift $\beta$ and the contrast may be controlled by varying the dispersion parameter $e_{disp}$. The oscillations are the result of the interference between the incident field whose frequency is fixed and the resonant part of the radiated field whose frequency is time dependent through its light-shift dependence. They represent thus a mapping of the light-shift in the time domain. An important remark can be done at this level. When the relative pulse duration varies, the resonant contribution changes. For instance, if at the entrance of the medium the propagating pulse is shorter than the driving one $\tau_{pd}<1$, the light-shift are important when the propagating pulse acts and the resonant part of the coherence (21) vanishes (because $\int_{-\infty}^{-T_0} f_p(Z,T')dT'\simeq 0$). The medium becomes transparent to the propagating pulse in this case and the propagating pulse is transmitted with almost no distortion. In the opposite case, when $\tau_{pd}>1$, the propagating pulse excites resonantly the system before the driving pulse freezes this interaction. The corresponding radiation induced by this non vanishing contribution interferes with the incident field to give rise to the observed oscillations.

**III-2 Three-level ladder system situation**

A similar phenomenon can be observed in a three-level system with collinear driving and propagating pulses acting on different energy levels ($\vec{k}_p // \vec{k}_d$). Consider a three-level ladder system with two excited states being $\left|1\right\rangle$ and $\left|2\right\rangle$ and coupled by the same driving pulse as in section II (fig. 3). In addition consider that state $\left|1\right\rangle$ is coupled resonantly to the ground state $\left|0\right\rangle$ by the weak propagating pulse whose expression is given in relations (10, 15). We write the wave function in adiabatic basis as:

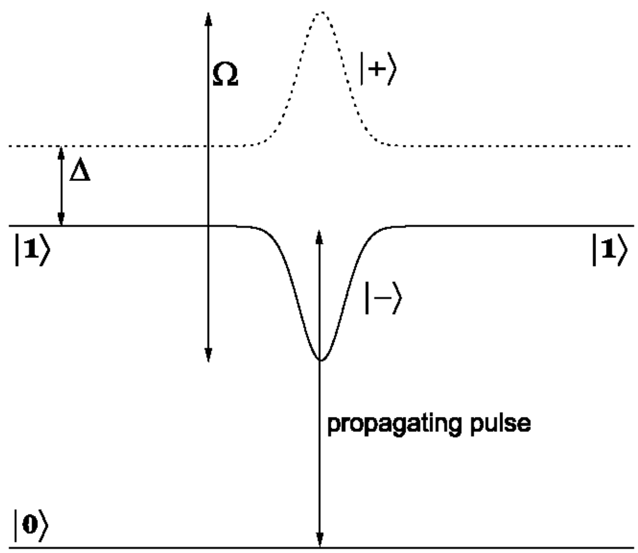


**Figure 3.** Schematic representation of the three-level system in the adiabatic representation associated with the driving pulse. Only, state |−⟩ can be efficiently coupled to the ground level.

$$|\psi\rangle(Z,T) = a_0(Z,T)|0\rangle + \alpha_-(Z,T)|-\rangle + \alpha_+(Z,T)|+\rangle \tag{23}$$

The equation for the evolution of amplitudes is given by:

$$i\partial_T \begin{pmatrix} a_0 \\ \alpha_- \\ \alpha_+ \end{pmatrix} = \frac{1}{2}\begin{pmatrix} 0 & -\frac{\theta_p f_p^*}{\tau_{pd}}\cos\phi & \frac{\theta_p f_p^*}{\tau_{pd}}\sin\phi \\ -\frac{\theta_p f_p}{\tau_{pd}}\cos\phi & (\Delta-\Omega) & 2i\partial_T\phi \\ \frac{\theta_p f_p}{\tau_{pd}}\sin\phi & -2i\partial_T\phi & (\Delta+\Omega) \end{pmatrix}\begin{pmatrix} a_0 \\ \alpha_- \\ \alpha_+ \end{pmatrix} \tag{24}$$

With $\theta_p$ now given as $\theta_p = \mu_{01}\,\varepsilon_{0p}\tau_p/\hbar$. It can be seen that the ground state is a stationary state and is coupled by the propagating pulse to the two dressed states. The coupling is influenced by the driven pulse through the sine and cosine factors. We will assume next that condition (9) is fulfilled so adiabatic evolution is obtained on the upper transition. The propagating pulse obeys to a propagation equation similar to equation (19) with the radiating coherence in the direction of the weak field $\rho_p = a_0^* a_1$. From (7-a), we obtain:

$$\rho_p = a_0^*\alpha_- \cos\phi - a_0^*\alpha_+ \sin\phi \tag{25}$$

The first part represents the contribution of transition $|0\rangle \to |-\rangle$ and the second part the contribution of transition $|0\rangle \to |+\rangle$. Because the detuning $\Delta$ is large and if initially all population is in the ground state, the population can be transferred efficiently only to state $|-\rangle$ that is resonant with the propagating field before the application of the driving pulse. Within these conditions, $\alpha_+ \simeq 0$, $a_0 \simeq 1$. We introduce now times $-T_0'$ and $T_0'$ solutions of the equation $(\Omega-\Delta)/2 = \tau_{pd}^{-1}$. The time interval between these two times represents again the time during which the light-shift induced by the driving pulse is important (larger than the propagating pulse bandwidth). The dominant contribution to the amplitude $\alpha_-$ is given by resonant part corresponding to the application of the propagating pulse out of this time interval. Using equations 24-25, the coherence $\rho_p$ for time T with $-T_0' \le T \le T_0'$ can be approximated by:

$$\rho_p \simeq \frac{i\theta_p}{2\tau_{pd}} \cos\phi \, e^{-i\Delta T} \, e^{-i\int_{-\infty}^{T} \frac{(\Omega-\Delta)}{2} dT'} \int_{-\infty}^{-T_0'} f_p(Z,T')dT' \qquad (26)$$

The transmitted intensity $I_p(1,T) = \left|\varepsilon_{0p}\, f_p(1,T)\right|^2$ can be approximated for $-T_0' \le T \le T_0'$ at the lowest order of the dispersion parameter by the following expression:

$$I_p(1,T) \simeq I_{0p}\left[\left|f_p(0,T)\right|^2 - e_{disp}\cos\phi(T)\left(\cos\left(\int_{-\infty}^{T}\frac{(\Omega-\Delta)}{2}dT' - \beta\right)\right)\left|\int_{-\infty}^{-T_0'} f_p(0,T')d\left(\frac{T'}{\tau_{pd}}\right)\right|\right] \qquad (27)$$

This expression is similar to that obtained in the case of a two-level atom (equation 22). Again we see that the transmitted pulse intensity is modulated with an interference which depends on the light-shifts. Two differences appear here: first, the light-shift in the three-level system is half of that of two-level system (with the same dipole moment). Secondly, the dependence of the modulation depends on $\cos^2\phi$ in the first case and on $\cos\phi$ in the second one. These differences occur because only the excited state is connected by the driving pulse in the first case while both the ground and excited states are connected by the driving pulse in the second case and are sensitive to the light-shifts effects.

### III-3 Numerical results and experimental implementation

In fig. 4, we represent in the case of a two-level system, the propagating pulse at the entrance of the medium (dotted line), the transmitted pulse in the absence (dashed line) and in the presence of the driving pulse (solid line).

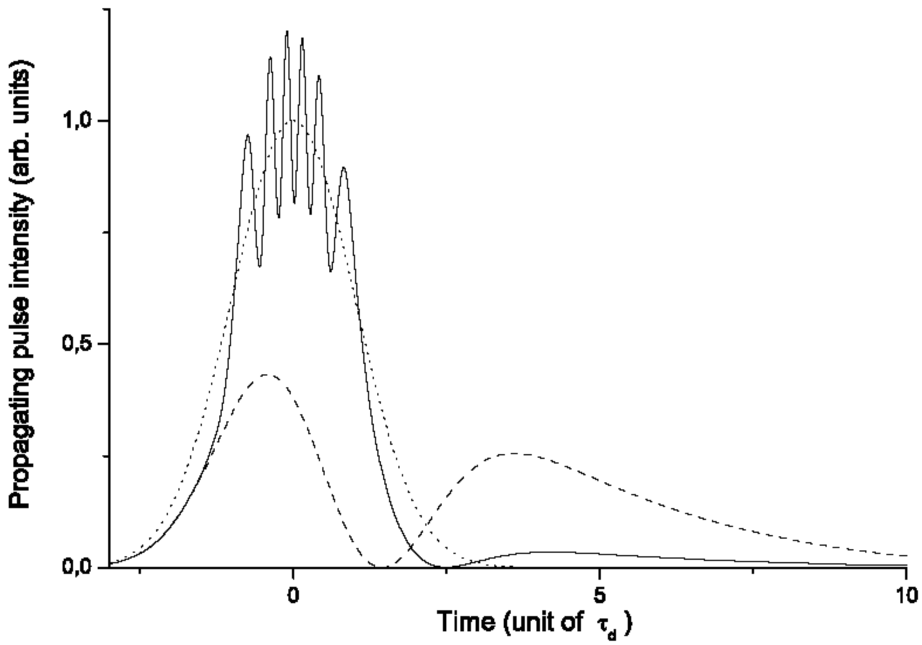


**Figure 4.** Two-level system. Intensity envelope of the propagating pulse at the entrance of the medium (dotted line), and at the exit, in the absence (dashed line) and the presence (solid line) of the driving pulse. Here, $\theta_d = 60,\ \theta_p = 0.01, \Delta = 10, \beta = 0\ \ \tau_{pd} = 2$ and $e_{disp} = 1$.

The parameters are $\tau_{pd} = 2$, $\theta_d = 60$, $\Delta = 10$, $\beta = 0$, $e_{disp}$ = 1 and $\theta_p = 0.01$. These curves are obtained by solving numerically the Maxwell-Schrödinger equations of the two-level system interacting with both pulses. Two features characterize the action of the driving pulse. First, the oscillations at long time scale, due to the atomic dispersion are strongly reduced because the driving pulse significantly reduces the coherence amplitude and thus the radiated intensity. The energy is concentrated in the central peak (pulse envelope at $T \simeq 0$) as a result of it. Secondly, tiny oscillations appear in this central peak. The combined action of the driving pulse and the propagation effects thus result in the transformation of a smooth pulse at the entrance of the medium into a modulated pulse at the exit. An important aspect of the interaction is the enrichment of the spectrum of the propagating pulse. We represent in fig. 5, in solid line the modification of the spectrum of the transmitted pulse for the same parameters as in fig. 4. We see that the spectrum contains new frequencies. They correspond to the light-shift induced on the transition $1 \to 4$. These frequencies belongs to a spectral band that spreads from $W_d$ to a maximum $W_d + \sqrt{\theta_d^2/\pi + \Delta^2}$ . Each frequency in this band appears twice in time giving rise to interference effects as shown in the figure.

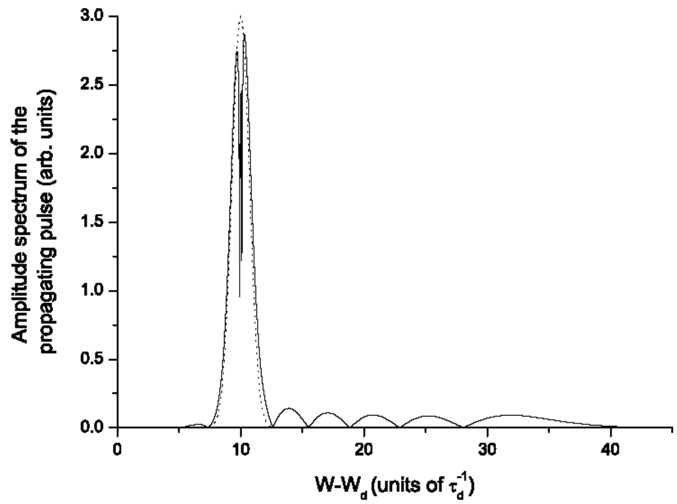


**Figure 5.** Two-level system. Amplitude spectrum of the propagating pulse, at the entrance (dotted line) and at the exit (solid line).The parameters are the same as in fig. (4).

For three-level system, similar behaviour is obtained for numerical results (fig. 6 and 7). We have chosen identical parameters to compare between this case and the two-level system situation. As said above, the difference lies in the temporal phase of interference. The propagating pulse experiences in the case of the three-level only half of the light-shifts induced by the driving pulse. Therefore, less oscillations appear in both temporal and spectral profiles of the transmitted pulse.

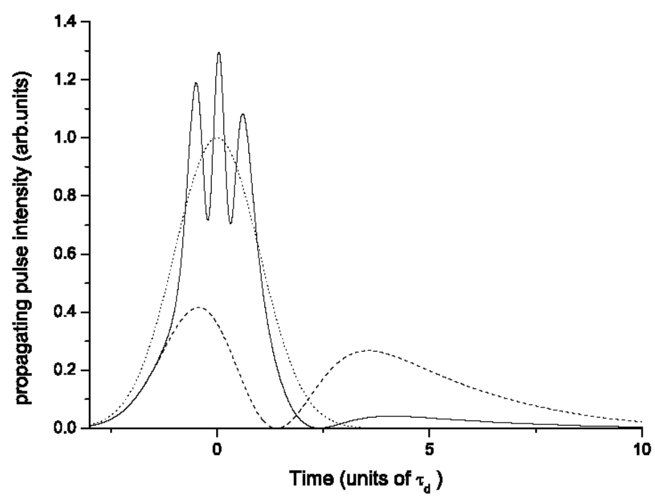


**Figure 6.** Three-level system. Intensity envelope of the propagating pulse at the entrance of the medium (dotted line), and at the exit, in the absence (dashed line) and the presence (solid line) of the driving pulse. Here, the parameters are the same as for the two-level system (fig. 4).

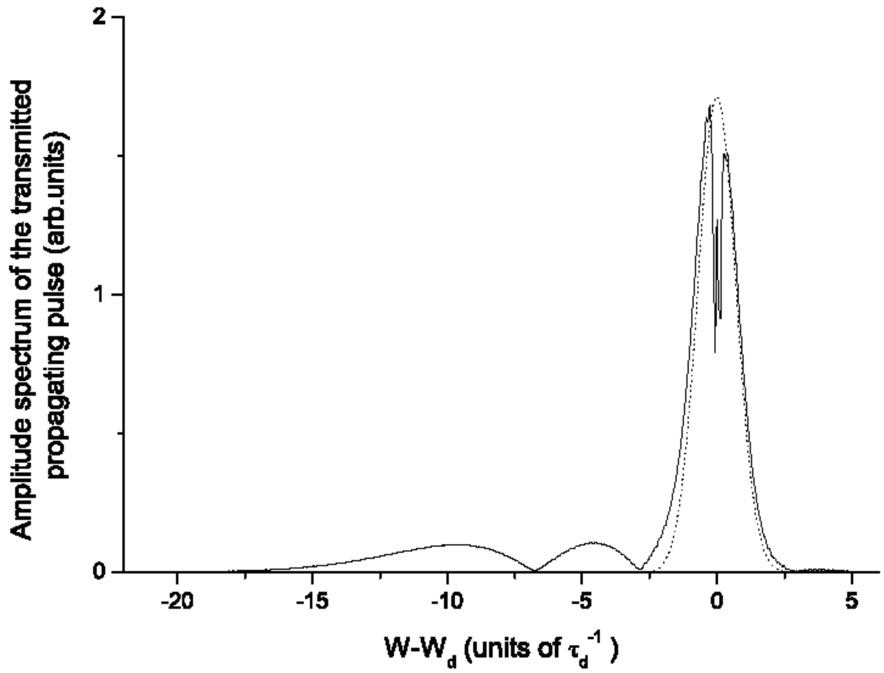


**Figure 7.** Three-level system. Amplitude spectrum of the propagating pulse, at the entrance (dotted line) and at the exit (solid line).The parameters are the same as in fig. (4).

The effects presented here can be observed for picosecond pulses in a two-level system with a large dipole moment, ensuring the induction of large light-shifts without ionising the atomic system. For instance in Rb atoms on the transition $5s\ ^2S_{1/2} \to 5p\ ^2P_{1/2}$ with $\lambda = 794.76\,nm$ and $\mu \simeq 1.7\,a.u$ and assuming Gaussian incident pulses with $\tau_d = 10\,ps$, a beam waist $w_0 = 2mm$ and an energy of $7.2\,\mu J$, we get $\theta_d = 60$. A wavelength detuning of 0.33 nm gives $\Delta = 10$. The peak intensity is then $I \simeq 3\times10^6 W/cm^2$, sufficiently low to avoid direct ionisation or multiphoton processes in this system. The optical depths used in the simulations presented in this paper can easily be reached. For instance for a cell with a length $L = 1cm$ (smaller than the Rayleigh length $z_0 = \frac{\pi w_0^2}{\lambda} \simeq 15.8\,m$), we have $e_{disp} \simeq 1$ at $n = 1.1\times10^{13}\,at/cm^3$ . This atomic density is obtained for a cell temperature $T \simeq 100°C$. The Doppler width $\Delta_d = \sqrt{\frac{2k_B T}{m_{Rb}\lambda^2}}$ has a value of 0.37 *GHz*. The dipole dephasing time is then $\Delta_d^{-1}/\pi \simeq 0.86$ ns much larger than the typical characteristic time here ($\tau_d = 10\,ps$) ensuring that the energy deposition in the medium is small as assumed in this paper.

For a three-level system, experimental observation of these effects may be possible for instance in lead $^{208}Pb$ with the transitions $6s^2 6p^2\ {}^3P_0 \rightarrow 6s^2 6p7s\ {}^3P_1$ (propagating pulse) with $\lambda \simeq 283nm$, $gf = 0.2$ and the transition $6s^2 6p7s\ {}^3P_1^0 \rightarrow 6s^2 6p7p\ {}^3D_1$ (driving field) with $\lambda \simeq 1065nm$, $gf = 0.98$. For our proposal, the experiment may be carried out in the picosecond regime using Ti: Sapphire mode-locked lasers. The tunability of these lasers makes it easy to find the adiabaticity condition (e.g a detuning around 1 nm). Assuming 3 nJ pulses, $\tau_2 \simeq 20ps$ and a beam waist $w_0 \simeq 20\mu m$, we have $\theta_2 \simeq 55$. The peak intensity is then $I = 1.5 \times 10^7$ $W/cm^2$, sufficiently low to avoid multiphoton processes in this system. Using a thin cell with L=1 mm for instance, we have $e_{disp} \simeq 1$ at $N = 2\times10^{13} at/cm^3$. Detection of the intensity profile of the 283 nm can be performed by cross-correlation in a gas.

# IV. Analogy with chirped pulse propagation

The situation studied here presents some analogy with the case of the propagation of chirped pulses where the interference between the incident and radiated fields reveals the sweeping of instantaneous frequency of the chirped pulse [10,14] whereas the situation discussed in §III reveals the sweeping of the atomic resonance frequency [8, 9]. We consider an ultra-short laser pulse (electric field given by relation 10) that propagates through the assembly of two-level systems. The envelope of the electric field at the entrance of the medium expresses as follows:

$$f_p(t,0) = \frac{1}{\sqrt{\pi}} e^{-(t/\tau)^2} e^{-i\delta t^2} \tag{28}$$

with $\tau = \tau_p \sqrt{1 + 4\frac{\phi_0^{"2}}{\tau_p^4}}$ and $\delta = \frac{2\phi_0^{"}}{\tau_p^4\left(1 + 4\phi_0^{"2}/\tau_p^4\right)}$ representing the pulse duration and the frequency sweeping in time respectively. Here $\phi_0^{"} = d^2\phi/d\omega^2$ represents the positive quadratic phase dispersion responsible for the group velocity dispersion and $\tau_p^{-1}$ represents the spectrum bandwidth. We consider only the case of highly chirped pulses $\phi_0^{"} >> \tau_p^2$ for which $\tau \simeq 2\phi_0^{"}/\tau_p$, $\delta \simeq 1/2\phi_0^{"}$ and we restrict our study to the weak field regime. During propagation, the temporal pulse profile is distorted. The envelope of the electric field at the exit of the medium ($Z = 1$) can be written as the sum of the incident field and the radiated field (equation 12, 13). In the following, we focus on the particular case of a weakly dense medium such as $e_{disp} << \sqrt{\tau_p/\tau}$. For such dispersion parameter, a chirped laser pulse undergoes strong distortion while a Fourier limited pulse with the same spectrum bandwidth undergoes little distortion. The behaviour of the radiated field can be understood more clearly once limiting the observation to a time interval $\sqrt{\delta^{-1}} << t << \tau, \tau_p / e_{disp}$. We obtain then $R(t') \simeq -e_{disp} / \tau_p$, $f_p(t,0) \simeq \frac{1}{\sqrt{\pi}} e^{-i\delta t^2}$ and the envelope of the radiated field might be approximated simply by :

$$f_{rad} \simeq -e_{disp} \sqrt{\tau / \tau_p} \, e^{-i\pi/4} \tag{29}$$

First, in the limit of our approximation ($e_{disp} << \sqrt{\tau_p / \tau}$), the radiated field is smaller than the incident one, that implies that the atoms feel essentially the interaction of the incident field. This last one can be seen as a quasi monochromatic field whose frequency sweeps in time from the lower to the higher part of the spectrum. The instantaneous frequency varies linearly in time as $\omega_{inst} = \omega_0 + 2\delta t$. As a result, the atom begins to radiate efficiently once the instantaneous frequency coincides with the atomic frequency, namely on a time interval $\pm\sqrt{\delta^{-1}}$ located around $t \simeq 0$. Then the atomic dipole radiates as long as damping effects are inefficient. This contribution that builds up on a time scale of the order of $2\sqrt{\delta^{-1}}$ represents thus the resonant and dominant contribution.

The transmitted field is obtained by summing the incident field and the radiated field, $I_p(1,t) = \left|\varepsilon_{0p} \, f_p(1,t)\right|^2$. From equations (12, 13, 28, 29), we obtain for $\sqrt{\delta^{-1}} << t << \tau, \tau_p / e_{disp}$:

$$I_p(1,t) \simeq I_{op} \left(1 - 2\sqrt{\pi \tau / \tau_p} \, e_{disp} \cos\left(\delta t^2 - \pi/4\right)\right) \tag{30}$$

The transmitted intensity here reveals the quadratic phase of the incident chirped phase while the output intensity in §III reveals the phase of the light-shift induced by the driving pulse.

Fig. 8 shows the result of numerical simulations for the transmitted intensity of a chirped pulse. For negative times, the excitation of the two-level system is not resonant and the radiated field is not efficiently emitted. The transmitted field coincides with the incident one. For $t \simeq 0$, the chirped pulse excites the system that begins to radiate efficiently. The interference between the radiated field and the incident field leads for $t \geq 0$ to strong oscillations that reveal the quadratic phase of the incident chirped pulse.

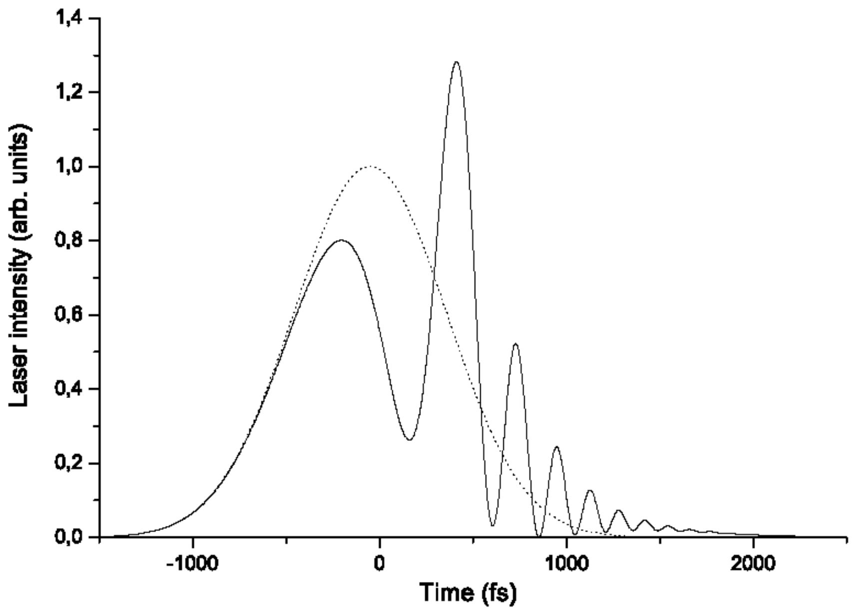


**Figure 8.** Laser intensity at the entrance (dotted line) and at the exit (solid line) of an ultrashort chirped pulse with $\tau_0 = 90\,fs, \phi_0^{''} = 3\times10^4\,fs^2$ propagating in an optically dense medium with $e_{disp} = 1$.

# V. Conclusion and perspectives

We have studied in this paper the reshaping effects that a weak resonant pulse experiences when propagating in an optically dense assembly of atoms driven by a strong non-resonant ultrashort pulse. We have considered two configurations: the first one corresponds to the situation where the two-pulse sequence excites the same two-level system and the second configuration corresponds to the case where the pulses connect different transitions of a three-level ladder system. Even if the theoretical analysis is different, the result remains the same in both cases. Moreover, the shape of the transmitted pulse depends strongly on its duration with respect to the driving pulse. When the driving pulse is shorter, an important modulation is exhibited in the temporal profile of the weak pulse that maps out the transient light-shift induced by the driving pulse while the long time range tail is significantly reduced. In the opposite case, the light-shifts prevent the propagating pulse from interacting efficiently with the atomic system and the propagating pulse is transmitted with almost no distortion. The medium becomes transparent to the propagating pulse in this latter case. An analogy with the propagation of an ultrashort chirped pulse through an optically dense medium of two-level atom has been presented to exhibit more clearly the role of the light-shift on the radiated field. They induce a sweeping of the frequency of the radiated field like the chirp is responsible for the sweeping of the central laser frequency. The consequence is the same in both cases since the associated phase is mapped out in the transmitted propagating pulse.

In addition to the possibility given by this method to measure the light-shifts by recording the transmitted profile of the propagating pulse, these results show that an optically dense atomic system driven by a strong pulse can be used as a "pulse-shaper" that can modify the temporal shape of an ultrashort weak pulse. Application of this method is restricted to the shaping we can obtain through the phase induced by light-shifts. Complex forms can be generated by modifying the envelope of the driving pulse to obtain different light-shift and thus different output shaped pulses. The crucial advantage of this method is the possibility to shape a pulse at a characteristic time smaller than its Fourier transform time duration which is not possible with standard techniques since it requires the creation of new frequencies. This method also exhibits many advantages over conventional devices [1]. These latter don't operate in the VUV domain, and are indirect methods since the temporal modifications are obtained by acting in the spectral domain. Strong limitations arise because of the spectral resolution of the devices that limits generally the shaping to very short pulses and the temporal resolution lies in the picosecond range making them inadequate to shape pulses in inertial fusion experiments [15]. The method presented here allows a direct shaping in the time domain avoiding the above problems and represent an ideal complement for the conventional methods while operating in the picosecond range and/or using UV pulses. The modulation can be varied through the laser and medium characteristics (optical depth, driving pulse intensity, relative pulse durations) providing a large range of control parameters.

# References

1. A. M. Weiner, "Femtosecond pulse shaping using spatial light modulators," Rev. Sci. Instrum. **71**, 1929 (2000).
2. D. Zeidler, T. Hornung, D. Proch, M. Motzkus, "Adaptive compression of tunable pulses from a non-collinear type OPA to below 20fs by feedback-controlled pulse shaping," Appl. Phys. B **70**, 125 (2000).
3. R. Bartels, S. Backus, E. Zeek, L. Misoguti, G. Vdovin, I. P. Christov, M. M. Murnane, H. C. Kapteyn, "Shaped-pulse optimization of coherent emission of high-harmonic soft X-rays " Nature **406**, 164 (2000).
4. A. Assion, T. Baumert, M. Bergt, T. Brixner, B. Kiefer, V. Seyfried, M. Strehle, G. Gerber, "Control of Chemical Reactions by Feedback-Optimized Phase-Shaped Femtosecond Laser Pulses" Science **282**, 919 (1998)
5. M. M. Wefers, K. A. Nelson, "Analysis of programmable ultrashort waveform generation using liquid-crystal spatial light modulators," J. Opt. Soc. Am. B **12**, 1343 (1995).
6. J. X. Tull, M. A. Dugan, W. S. Warren, "High Resolution, Ultrafast Laser Pulse Shaping and its Applications," Adv. Magn. Opt. Reson. **20**, 1 (1997).

7. E. Zeek, K. Maginnis, S. Backus, U. Russek, M. Murnane, G. Mourou, H. Kapteyn, G. Vdovin, “Pulse compression by use of deformable mirrors” Optics Letters **24**, 493 (1999)
8. J. C. Delagnes and M. A. Bouchene, “Effect of a transient light-shift on the propagation of an ultrashort pulse in an atomic resonant medium‘’, Phys. Rev. A **69**, 63813 (2004)
9. J. C. Delagnes, F. A. Hashmi and M. A. Bouchene, "Spectral and temporal modifications of a weak resonant ultrashort pulse propagating in a two-level system driven by a strong non-resonant pulse" Phys. Rev. A, **74**, 053822 (2006)
10. J. C. Delagnes, V. Blanchet and M. A. Bouchene, ‘’ Role of the radiated field in the propagation of an ultra-short chirped pulse‘’, Optics Communications **227,** 125 (2003)
11. P. R. Berman, L. Yan, K-H. Chiam, "Nonadiabatic transitions in a two-level quantum system: Pulse-shape dependence of the transition probability for a two-level atom driven by a pulsed radiation field" Phys. Rev. A **57**, 79 (1998)
12. P. W. Milonni and J. H. Eberly, Lasers (Wiley, New York, 1988), p. 251-252
13. M. A. Bouchene, “Phase Control of Dispersion Effects for an Ultrashort Pulse-train Propagating in a Resonant Medium”, Phys. Rev. A **66**, 065801 (2002)
14. J. E. Rothenberg and D. Grischkowsky, “Measurement of the phase of a frequency-swept ultrashort optical pulse,” J. Opt. Soc. Am. B **2**, 626 (1985).
15. X. Ribeyre, C. Rouyer, F. Raoult D. Husson, C. Sauteret, A. Migus “All-optical programmable shaping of narrow-band nanosecond pulses with picosecond accuracy by use of adapted chirps and quadratic nonlinearities **“**, Optics Letters **26,** 1173 (2001)