Effect of a transient light shift on the propagation of an ultrashort pulse in a resonant atomic medium

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An ultrashort weak pulse that propagates resonantly on the $|a\rangle \rightarrow |b\rangle$ transition of a three-level system $(|a\rangle, |b\rangle, |c\rangle)$ experiences reshaping effects on a time scale longer than its duration. When a strong field is applied (nonresonantly) on the $|b\rangle \rightarrow |c\rangle$ transition, important light shifts are induced. We have theoretically studied the novel behavior of both the medium and weak propagating laser pulses in such conditions. For the transmitted field profile, drastic changes occur: at long times, efficient reduction of the distortion is obtained, while strong oscillations appear for short times, mapping out the dynamical light shift. These shaping effects, which are accompanied by new features in the spectrum for the propagating pulse, are interpreted as the result of the interference between incident field and induced dipole radiated field, whose frequency sweeps in time because of the induced light shift.

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I. INTRODUCTION

The interaction of a pair of weak and strong laser pulses with three-level systems leads to a great variety of spectacular phenomena. Many of them deal with the case where the strong pulse is sufficiently long to bring the system to a stationary state while the weak pulse is acting. These situations can thus be viewed as pump-probe experiments where the weak field probes the time-invariant linear response of the system modified by the strong pulse. This situation was explored, for instance, in saturation spectroscopy [1], optical-field-induced birefringence [2], and electromagnetically induced transparency [3]. In this latter case, 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 [4]. When the action of the strong pulse is time dependent, the weak pulse probes the transient dynamics induced by the strong pulse and the time invariance of the response function is no longer satisfied. When the strong pulse dynamically Stark shift a bound state, Ramsey-like fringes appear in the absorption linewidth of the probe pulse that connect this state to an unperturbed one [5]. It may also be possible to extract the dynamic shift if the probe pulse is at least as short as the half-cycle period of the strong pulse [6]. When the strong pulse has a time duration shorter than the weak pulse, both the time invariance property and the pump-probe picture break down and transient effects dominate the dynamics during the action of the weak pulse. We investigate here the transient effects experienced by a weak pulse that propagates through an atomic medium driven by a strong laser pulse (Fig. 1). The study of propagation effects in resonant media is a powerful tool to explore the dynamics in both atomic and molecular systems [7,8]. On the other hand, the possible application of ultrashort-pulse shaping in these media may be a alternative solution to conventional pulse shapers in the spectral domain where these are no longer effective.

In this paper, the weak pulse is designated as the propagating pulse and interacts resonantly with the atom on the $|a\rangle$-$|b\rangle$ transition. The strong pulse, designated as the driving pulse, is applied on the $|b\rangle$-$|c\rangle$ transition. We restrict the study to the case where the driving pulse is off resonance such that adiabatic evolution is expected on the $|b\rangle$-$|c\rangle$ transition and we assume that its time duration is smaller but comparable to that of the propagating pulse. In the case where the driving pulse is longer in time than the probe pulse, and the probe pulse is strong, the interaction leads to the coherent population return phenomena (CPR) where all the population is adiabatically transferred to the excited state and then back to the ground level [9]. In our configuration, we will show that transient effects lead to new reshaping effects of the propagating field where two kinds of interference are involved. The first type of interference takes place in atomic population and in the radiated field as well. This kind of interference results from the superposition of the on- and off-resonant excitation amplitudes of the excited states. The second type deals with the interference of the incident and radiated fields. Substantial modification of the propagating pulse profile is then obtained. First, strong oscillations appear mapping out the Stark-shift phase on the transmitted pulse. Second, an efficient reduction of the radiated field is

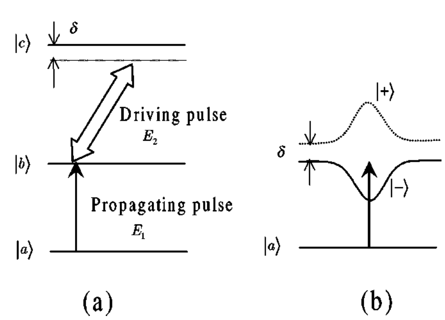

FIG. 1. (a) Schematic representation of the three-level system in the bare-state picture. (b) The equivalent scheme in the dressed-states picture of the strong driving pulse.

realized, making the distortion of the transmitted pulse strongly reduced for long times.

II. THEORETICAL MODEL

The electric field of the propagating pulse is expressed as $E_1=[\varepsilon_{01}f_1(z,t)e^{-i(\omega_1 t-k_1 z)}+\text{c.c.}]/2$, where $f_1(z,t)$ is the envelope, expressed at the boundary entrance of the medium as $f_1(0,t)\exp[-(t/\tau_1)^2]$. When the optical medium is thick, the pulse experiences strong reshaping effects while propagating. The temporal distortion of the pulse is the consequence of two propagation laws that have to be obeyed by the optical pulse [10]. First, the pulse is short. The spectrum width τ_1^{-1} is larger than the absorption line given by the Doppler width, so the majority of the initial energy is transmitted. The second propagation law concerns the MacCall-Hahn area theorem, which states that in the limit of the weak-field regime, the pulse area $\propto\int f_1(t,z)dt$ decreases exponentially with the propagation distance z. The pulse envelope thus develops an oscillatory temporal structure to satisfy both conditions. Substantial energy is then released in the long decreasing tail of the laser pulse. We study in the following the changes induced on the propagating pulse by the action of a strong pulse with electric field $E_2=(\varepsilon_{02}f_2(z,t)e^{-i(\omega_2 t-k_2 z)}+\text{c.c.})/2$, where $f_2(0,t)=\exp[-(t/\tau_2)^2]$ and that drives the transition $|b\rangle$-$|c\rangle$ (with eigenfrequency ω_{bc}). This pulse has a time duration smaller but comparable to that of the propagating pulse. We assume that substantial detuning on the $|b\rangle$-$|c\rangle$ transition and the small population transfer to the intermediate state by the weak-propagating field prevents any alteration of the strong field by the interaction with the atomic medium [i.e., $f_2(z,t)\simeq f_2(0,t)$]. The total wave function of the system is $|\psi\rangle=a(z,t)|a\rangle+b(z,t)e^{-i\omega_1 t}|b\rangle+c(z,t)e^{-i(\omega_1+\omega_2)t}|c\rangle$. The amplitudes a,b, and c and the propagating field envelope f_1 evolve accordingly to the Maxwell-Schrödinger equations which give using the new reduced variables $T=(t-z/c)/\tau_1$ and $Z=z/l$ (l is the length of the sample):

$$i\frac{\partial}{\partial T}\begin{pmatrix}a\\ b\\ c\end{pmatrix}=-\frac{1}{2}\begin{pmatrix}0 & \theta_1 f_1^* & 0\\ \theta_1 f_1 & 0 & \theta_2 f_2\tau_{12}\\ 0 & \theta_2 f_2\gamma_{12} & -2\delta\tau_{12}\end{pmatrix}\begin{pmatrix}a\\ b\\ c\end{pmatrix} \tag{1}$$

and

$$\frac{\partial}{\partial Z}f_1=i\frac{e_{\text{disp}}}{\theta_1}a^*b, \tag{2}$$

where $\theta_1=[(\mu_{ab}\varepsilon_{01})/\hbar]\tau_1$ and $\theta_2=[(\mu_{bc}\varepsilon_{02})/\hbar]\tau_2$ are parameters that characterize the intensity strength for the propagating and the driving field, respectively, $\tau_{12}=\tau_1/\tau_2$, and $\delta=(\omega_{bc}-\omega_2)\tau_2$ represents the detuning of the driving field. The dimensionless coefficient $e_{\text{disp}}=(N\mu_{ab}^2\omega_1/2c\varepsilon_0\hbar)l\ \tau_1$ characterizes the severity of dispersion effects on the $|a\rangle$-$|b\rangle$ transition and depends on the atomic density N. Moreover, this quantity can be related to the better known optical depth parameter $\alpha_0\ l$ (α_0 is the absorption coefficient at resonance and l is the length of the sample) by $e_{\text{disp}}=\alpha_0 l\Delta_d\tau_2$, where Δ_d is the Doppler width. Here $\alpha_0 l\Delta_d$ represents the spectral domain (around the resonance) over which the dispersion affects the spectral phase of the incident pulse [11]. Thus $e_{\text{disp}}=\alpha_0 l\Delta_d\tau_2$ can be interpreted as the ratio between this quantity and the spectral bandwidth of the pulse. Therefore $e_{\text{disp}}\geqslant 1$ means that dispersion alters the phase of all the spectral components of the incident pulse and so propagation effects cannot be neglected.

Equations (1) and (2) constitute a complete set of partial differential equations that can be solved numerically. An alternative representation uses dressed states where the action of the driving pulse can be more clearly exhibited. It can be obtained after performing the unitary transformation

$$\begin{pmatrix}a_-\\ a_+\end{pmatrix}=\begin{pmatrix}\cos\phi & \sin\phi\\ -\sin\phi & \cos\phi\end{pmatrix}\begin{pmatrix}b\\ c\end{pmatrix}, \tag{3}$$

with

$$\tan[2\phi(T)]=\frac{\theta_2}{\delta}f_2(0,T). \tag{4}$$

The amplitudes a, a_+, and a_- satisfy the differential equations

$$i\frac{\partial}{\partial T}\begin{pmatrix}a\\ a_-\\ a_+\end{pmatrix}=\frac{1}{2}\begin{pmatrix}0 & -\theta_1 f_1^*\cos\phi & \theta_1 f_1^*\sin\phi\\ -\theta_1 f_1\cos\phi & \delta\tau_{12}[1-(\cos 2\phi)^{-1}] & 2i\ d\phi/dT\\ \theta_1 f_1^*\sin\phi & -2i\ d\phi/dT & \delta\tau_{12}[1+(\cos 2\phi)^{-1}]\end{pmatrix}\begin{pmatrix}a\\ a_-\\ a_+\end{pmatrix}. \tag{5}$$

In this representation the ground state appears as a stationary state coupled by the propagating pulse to the two dressed states. The coupling is slightly influenced by the driven pulse through the sine and cosine terms. The dressed states are separated by $\delta\tau_{12}(\cos 2\phi)^{-1}$ through the ac stark splitting or Autler-Townes effect [Fig. 1(b)] The coupling between these two states is performed by the derivative of the mixing angle. Adiabatic evolution of the system occurs if the coupling $2|d\phi/dT|$ is weaker than the ac Stark splitting $\delta\tau_{12}(\cos 2\phi)^{-1}$. From Eq. (5), the condition is expressed as

$$\left| \frac{\theta_2 \dfrac{df_2}{dT} \delta\tau_{12}}{(\theta_2^2 f_2^2 + \delta^2)^{3/2}} \right| \ll 1 \tag{6}$$

and may be verified for large detuning and/or strong pulses. In this case, using Eq. (5) and the fact that the transition $|a\rangle$-$|b\rangle$ is weakly driven ($a \simeq 1$), the amplitudes a_- and a_+ are given by

$$a_-(Z,T) \simeq \frac{i\theta_1}{2} \int_{-\infty}^{T} f_1(Z,T') \cos\,\phi(T') \times \exp\left(-i\int_{T'}^{T} \omega_- dT''\right) dT', \tag{7a}$$

$$a_+(Z,T) \simeq -\frac{i\theta_1}{2} \int_{-\infty}^{T} f_1(Z,T') \sin\,\phi(T') \times \exp\left(-i\int_{T'}^{T} \omega_+ dT''\right) dT', \tag{7b}$$

where $\omega_{\pm}(T) = \delta\tau_{12}\{1 \pm [\cos 2\phi(T)]^{-1}\}$ represent the eigenfrequencies of the adiabatic states (in units of τ_1^{-1}). For large detuning such as $\delta\tau_{12} \gg 1$ and because the propagating pulse is resonant with the adiabatic state $|-\rangle$ at the beginning of the action of the pumping pulse, the population is efficiently transferred solely to this state and so $a_+ \simeq 0$. In the next section, we show the results of the numerical resolution of the differential equations (1) and (2), and discuss the underlying physics for the behavior of the excited-state population and the radiated and transmitted fields.

III. RESULTS AND DISCUSSION

We set, throughout all this section, $\theta_1 = 0.15$, $\tau_{12} = 2$, and $e_{\text{disp}} = 1$. We also choose the detuning for the pumping pulse to be $\delta = 20$. Condition (6) is fulfilled and adiabatic evolution of the $|b\rangle$-$|c\rangle$ transition is obtained for the whole range of the considered strengths of the driving pulse. The temporal profiles of the populations in the excited states $|b\rangle$ and $|c\rangle$ at the boundary entrance of the medium ($z=0$) are plotted in Fig. 2 for two values of the strength parameter of the driving pulse. In (a), $\theta_2 = 0$, the $|c\rangle$ state is not populated while the population of state $|b\rangle$ grows to an asymptotic value on a time scale of τ_1. In (b), $\theta_2 = 60$, the population in state $|b\rangle$ increases initially under the action of the propagating pulse. When the strong pumping pulse acts, population is created in state $|c\rangle$ while the $|b\rangle$ state population stops to grow. During all this period, in both $|b\rangle$ and $|c\rangle$ the population is modulated. These oscillations are *in phase* and thus cannot be interpreted as Rabi oscillations induced by the strong field on the $|b\rangle \rightarrow |c\rangle$ transition. Note also the drastic change in the population level between cases (a) and (b). In this latter, when the driving pulse is terminated, the population in state $|c\rangle$ vanishes while the population of the $|b\rangle$ state increases again. These interesting features can be explained in the terms of the dressed-states representation when considering the adiabatic state $|-\rangle$ involved in the interaction process.

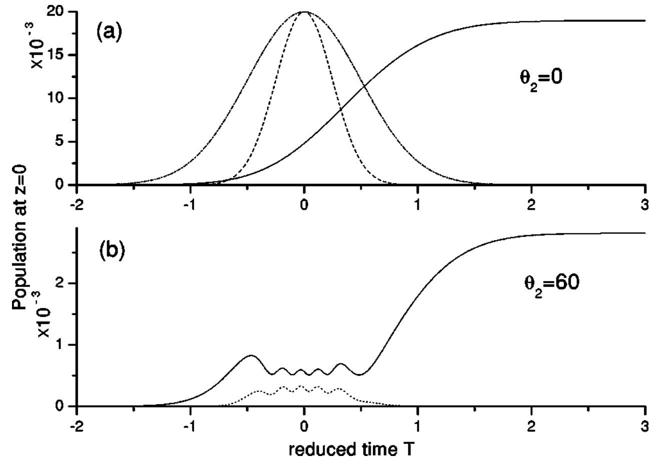

FIG. 2. Temporal evolution of the population in the excited states $|b\rangle$ (solid line) and $|c\rangle$ (dotted line) for two strengths of the pumping pulse: (a) $\theta_2 = 0$ and (b) $\theta_2 = 60$. We also represent in (a) the envelope of the propagating (dot-dashed curve) and driving (dashed curve) pulses with arbitrary units. See text for the values of the other parameters.

Let us consider the solutions $-T_0$, and T_0 of the equation $\omega_-(T) = 1$. The interval between these two times represents the time during which the light shift induced by the strong driving pulse is important and we assume for simplicity that the action of the driving field is nearly restricted to this time interval which is true for large values of θ_2. We designated the adiabatic-state amplitudes in region of $-\infty_0 < T \leq -T_0$, $-T_0 < T < T_0$, and $T_0 \leq T < 1$ by $\tilde{a}_-^{(1)} e(-i\int_{-\infty}^{T} \omega_- dT'')$, $\tilde{a}_-^{(2)} e(-i\int_{-\infty}^{T} \omega_- dT'')$, and $\tilde{a}_-^{(3)} e(-i\int_{-\infty}^{T} \omega_- dT'')$, respectively. Thus,

$$\tilde{a}_-^{(1)}(0,T) \simeq \frac{i\theta_1}{2} \left(\int_{-\infty}^{T} f_1(0,T') dT' \right), \tag{8a}$$

$$\tilde{a}_-^{(2)}(0,T) \simeq \tilde{a}_-^{(1)}(0,-T_0) + \frac{i\theta_1}{2} \left[\int_{-T_0}^{T} f_1(0,T') \cos\,\phi(T') \times \exp\left(+i\int_{-T_0}^{T'} \omega_- dT''\right) dT' \right], \tag{8b}$$

$$\tilde{a}_-^{(3)}(0,T) \simeq \tilde{a}_-^{(1)}(0,-T_0) + \frac{i\theta_1}{2} \left(\int_{T_0}^{T} f_1(0,T') dT' \right). \tag{8c}$$

For $-\infty_0 < T \leq -T_0$, prior to the action of the driving pulse, the states $|-\rangle$ identify with $|b\rangle$ and is populated by the propagating pulse. For $-T_0 < T < T_0$, the driving pulse induces an important light shift, preventing an increase in the population of state $|-\rangle$. However, even when weak, nonresonant excitation by the propagating pulse adds non-negligible contributions to the total amplitude of state $|-\rangle$. These contributions interfere with the amplitude brought at time $-T_0$ and oscillations are thus created in the time profile of the adiabatic-state population. Because the adiabatic state is a mixture of both $|b\rangle$ and $|c\rangle$ states, the interference process is also revealed through the population of these two states. Similar interfer-

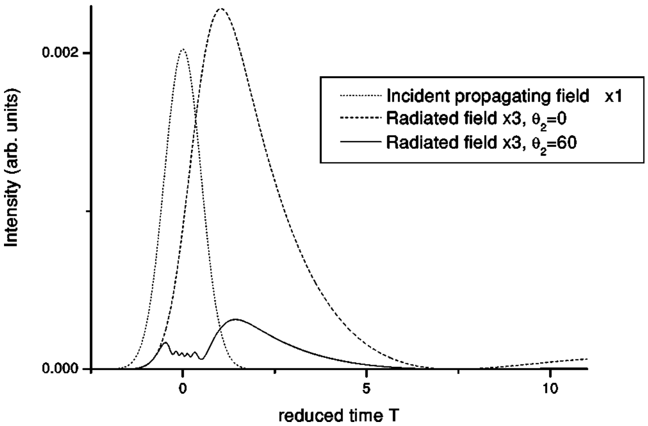

FIG. 3. Temporal behavior of the radiated field intensity at the output of the atomic medium for two different strengths of the pumping pulse. The laser and atomic parameters are the same as in Fig. 2.

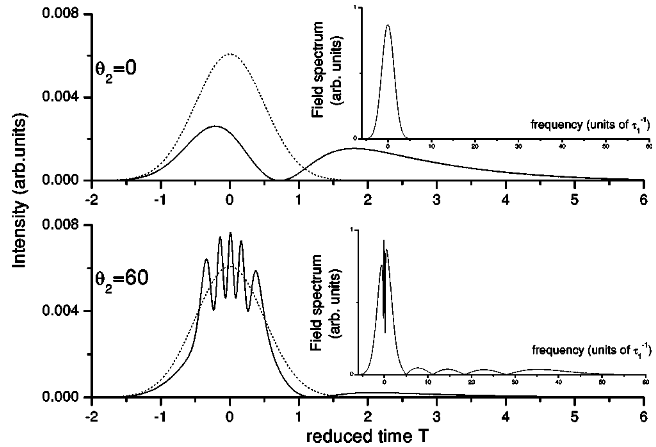

FIG. 4. Solid line: temporal behavior of the transmitted intensity for two different strengths of the pumping pulse. Dotted line: intensity of the incident pulse. The laser and atomic parameters are the same as in Figs. 2 and 3. The insets represent the modulus of the Fourier transform of the transmitted field for the corresponding situations.

ences appear also in two-level atoms where the laser frequency is swept instead of the atomic frequency [12]. These oscillations can also be interpreted as nonresonant Rabi oscillations *induced by the weak-propagating pulse* on the $|a\rangle \to |-\rangle$ transition. For $T_0 \leqslant T < 1$, the adiabatic state is no longer shifted and a new contribution is added to the residual amplitude, $\tilde{a}_-^{(2)}(0,T_0)$. The population increases again after the action of the driving pulse (i.e., $T \geqslant 1$). These features have a great impact on the distortion of the propagating pulse. The substantial decrease of the population in the $|b\rangle$ state when $\theta_2 \gg 1$ allows us to approximate at the lowest order the output radiated field envelope $f_{\mathrm{rad}}(T) = f_1(1,T) - f_1(0,T)$ by the expression [using relations (2), (3), and (7a)]

$$f_{\mathrm{rad}}(T) \simeq -\frac{e_{\mathrm{disp}}}{2}\cos\,\phi(T) b(0,T). \tag{9}$$

Except for the slowly varying cosine term [with $\cos\,\phi(\pm\infty = 1)$], the radiated intensity exhibits the same features as the $|b\rangle$-state population at the entrance boundary of the medium. Figure 3 confirms this result. Note here the substantial decrease of the radiated field intensity between the two cases where no strong driving pulse is present ($\theta_2 = 0$) and the situation where it acts.

The behavior of the total transmitted intensity that can be experimentally detected is shown in Fig. 4. It results from the superposition of the incident pulse and the radiated field. For $\theta_2 = 0$, the transmitted field is distorted and the long-time-scale oscillations observed are in accordance with the prevision of the McCall-Hahn theorem that holds in this case (see Sec. II). For $\theta_2 = 60$, different behavior occurs. Strong oscillations appear in the short-time range ($T < 1$), but the oscillations in the long-time range ($T \geqslant 1$) that are present with just the propagating pulse alone are strongly reduced. These effects can be understood by considering the process of interference between the incident and radiated field. For $T \geqslant 1$, only the radiated field is present and the intensity depends on the behavior of the population of the $|b\rangle$ state at the entrance boundary of the medium. This has been already discussed above; the level of energy spread in this region is low. In the short-time range, the oscillations result from the interference between the incident and radiated fields. This effect is different from the interference process that leads to the modulation of the exited state populations. For $-\infty < T \leqslant T_0$, because the nonresonant contributions are smaller than the resonant ones, the interference process between the radiated field and the incident field involves mainly the resonant contribution given in Eq. (8a). Using the relations (2), (3), (8a), and (9) and remembering that the radiated field has a much lower intensity than the incident one, it follows that the transmitted intensity $I(1,T) = |\varepsilon_{01} f_1(1,T)|^2$ is given by the following expression when $-\infty < T \leqslant T_0$:

$$I(1,T) = I_0\Bigg[|f_1|^2(0,T) + \Bigg|e_{\mathrm{disp}}\cos\,\phi(T) \times\left(\int_{-\infty}^{-T_0} f_1(0,T')\cos\,\phi(T')dT'\right)\Bigg| \times\cos\int_{-\infty}^{T}\omega_-(T')dT'\Bigg], \tag{10}$$

with $I_0 = \varepsilon_{01}^2$. After the end of the pumping pulse $T_0 \leqslant T < 1$, new resonant contributions are added, but the phase remains unchanged. The interference phase is given at time T by $\int_{-\infty}^{T}\omega_- dT'$, and the total number of oscillations is $\int_{-\infty}^{+\infty}\omega_- dT'/2\pi$. Numerical estimation of this integral gives 4.5 for $\theta_2 = 60$ in accordance with the number of oscillations observed in Fig. 4. An important feature is the dependence of the contrast of oscillations on the parameter e_{disp} that can be controlled through the atomic density N. The situation studied here presents some analogy to 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 whereas here the interference reveals the sweeping of the atomic resonance frequency [13]. We also report in the insets of Fig. 4 the modulus of the Fourier transform of the transmitted field. In the presence of the driving field, the spectral distribution is profoundly altered. New frequency bands appear and are the result of the cross-phase modulation induced by the strong pulse on the weak pulse. Moreover, the instantaneous frequency of the radiated field is given by $\omega_{-}(T)$ and sweeps across a spectral range of the order of $\delta\tau_{12}[1-(\cos 2\phi_{\max})^{-1}]/2 \simeq 45$, thus exceeding the spectral width of the propagation pulse. Because each frequency is involved twice during the application of the driving field, this gives rise to interference effects in the spectral domain as exhibited in the inset.

IV. CONCLUSION

In summary, we have studied how a transient off-resonant strong field modify the propagation of a weak resonant pulse in a three-level system. During the application of the strong field, two kinds of interference are involved in the process. The off- and on-resonant amplitude transfers from the fundamental to the excited states give rise to interference leading to the modulation of the populations of these states and the radiated field also. The interference between the incident and radiated fields leads to a strong modulation of the transmitted pulse that maps out the Stark shift induced by the strong pulse. Only the resonant part of the radiated field is involved in this interference process. At the end of the strong driving pulse, the substantial reduction of the population in the excited states lead to an efficient reduction of the long-time range distortion of the weak pulse.

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 $\lambda \simeq 283$ nm, $gf=0.2$ and the transition $6s^2 6p7s\ {}^3P_1^0 \rightarrow 6s^2 6p7p\ {}^3D_1$ (driving field) with $\lambda \simeq 1065$ nm, $gf=0.98$. This system has already been used for electromagnetic-induced transparency (EIT) experiments in the nanosecond regime [14]. For our proposal, the experiment may be carried out in the picosecond regime using Ti: sapphire mode-locked lasers that deliver 2×283 nm and 1065 nm pulses, respectively. The tunability of these lasers makes it easy to find the adiabaticity condition (e.g., a detuning around 1 nm). Assuming 3-nJ pulses, $\tau_2 \simeq 20$ ps, and a beam waist $w_0 \simeq 20\ \mu$m, we have $\theta_2 \simeq 55$. The peak intensity is then $I \simeq 1.5\times 10^7$ W/cm^2, sufficiently low to avoid multiphoton processes in this system. Using a thin cell ($l=1$ mm, for instance), we have $e_{\text{disp}} \simeq 1$ at $N=2\times 10^{13}$ at./cm^3. Detection of the intensity profile of the 283-nm laser pulse can be performed by cross correlation in a gas.

An enrichment of the transmitted pulse spectrum (cf. Fig. 4) is in line with these effects. It makes these kinds of modulations impossible to achieve with conventional pulse shapers [15] that are not able to amplify or create new frequency components. These results may be promising for the achievement of real-time modulation of ultrashort pulses controlled by both the intensity of the driving pulse and the atomic density of the medium.

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