Coherent Control of Light Shifts in an Atomic System: Modulation of the Medium Gain

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A sequence of two femtosecond coherent pulses—a strong π-polarized pulse and a weak σ-polarized pulse—excite the $S_{1/2}$-$P_{1/2}$ transition of atomic rubidium in an optically dense vapor. The σ pulse induces transitions between the adiabatic states with a coupling strength that is different for identically and oppositely light-shifted coupled states, and that can be modified by tuning the relative phase between the pulses. An efficient control of the medium gain for the σ pulse is experimentally demonstrated. It is shown to be the result of interference between the absorption and the stimulated emission paths for σ photons.

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The interaction of strong laser fields with matter causes light shifts that play an important role in many fields such as quantum optics, molecular physics, and chemistry. These effects induce dramatic changes in both the structure and the dynamics of the driven systems. Light shifts turn the single fluorescence line of a two-level atom into a Mollow triplet [1–3], they contribute to the appearance of electromagnetically induced transparency phenomena (EIT) [4] and are responsible for the Sisyphus mechanism of laser cooling [5]. These effects are strongly enhanced when laser pulses are used. Rapid adiabatic passage, stimulated Raman adiabatic passage, and Stark-chirped adiabatic passage are examples of effects where light shifts play a crucial role [6]. Furthermore, one can take advantage of light shifts to achieve pulse shaping in an atomic vapor [7]. In molecules, spectacular effects occur. For instance, light shifts cause light-induced potential (LIP) and new bound states appear [8]. Femtosecond pulses can be used to selectively open and close light-induced avoided crossings between two electronic potentials, thus modifying the final states [9,10]. Although light shifts prevail in many phenomena, only a few possibilities have been used to control their influence. This is generally done by controlling the laser intensity or the detuning when the interaction is not resonant [11]. In this Letter, we describe an experiment where we achieve coherent control of a light shift. We consider a double two-level system [Fig. 1(a)] where a strong femtosecond pulse drives the parallel transition and a weak resonant femtosecond pulse connects cross transitions. In the adiabatic representation [Fig. 1(b)], the dressed states are connected by the weak pulse with a coupling strength that differs depending on whether the states are identically or oppositely light shifted, and also depends on the relative phase of the optical waves. The control comes from adjusting this phase, which we achieve experimentally by varying the time delay between the pulses. Depending on the relative phase, the weak pulse can be amplified, or transmitted without any modification. As in previous coherent control experiments [12], the control here results from the interference between two excitation quantum paths, corresponding to absorption and to emission by the dressed atomic system of one photon from or to the weak field mode, respectively [Fig. 1(d)]. An original and interesting consequence of this mechanism is the period of the gain modulation as a function of pulse delay. It is half the optical transition period although only one-photon transitions are involved in the process.

We analyze now the interaction of the system described above with a pair of mutually coherent ultrashort pulses. This situation occurs when exciting the $S_{1/2}$-$P_{1/2}$ transition

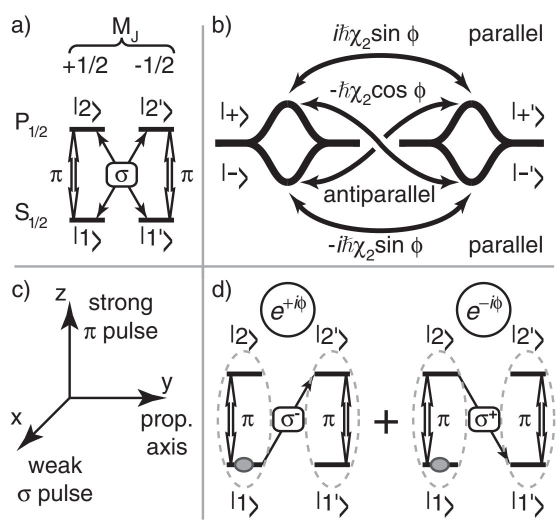

FIG. 1. (a) Energy levels and optical transitions involved in our system. (b) Energy levels in the adiabatic representation of the strong pulse. Parallel states and antiparallel states are coupled (by the σ pulse with Rabi frequency χ_2) differently through the phase-shift ϕ between the two pulses allowing coherent control of light-shift effects. (d) Representation of the interaction for atoms initially in state $|1\rangle$, as the result of interference between two quantum paths (with complex conjugate phases) involving absorption and emission of radiation.

of atomic rubidium with a pair of π and σ polarized pulses [Figs. 1(a)–1(c)]. The π-polarized pulse strongly couples resonantly the parallel states while the weak σ pulse couples resonantly the crossed states. The electric fields of the pulses propagating along the Oy axis in an optically dense medium with a delay τ, are expressed as $\vec{E}_\pi(y,t) = \vec{e}_z[\varepsilon_{01} f_1(y,t) e^{-i\omega t} + \text{c.c.}]$ and $\vec{E}_\sigma(y,t) = \vec{e}_x[\varepsilon_{02} f_2(y,t) e^{i\phi} e^{-i\omega t} + \text{c.c.}]$. Here $\phi = \omega\tau$ is the relative phase between the pulses, and t represents the local time ($t = t_{\text{lab}} - y/c$). We consider identical Gaussian pulses which nearly overlap temporally so that $f_1(y=0,t) = f_2(y=0,t) = \pi^{-1/2} e^{-t^2/\tau_0^2}$ with $\tau \ll \tau_0$ We explain now the interference effects that appear in such a situation by analyzing the behavior of the atomic wave function and the transmitted intensity of the σ pulse.

We focus first on the atomic dynamics at the entrance of the medium. The atoms are initially statistically distributed between the two ground levels $|1\rangle$ and $|1'\rangle$. From symmetry we need consider only those in state $|1\rangle$. Within the rotating wave approximation, the Hamiltonian of interaction due to the σ pulse can be split as $H_\sigma = H_{\sigma^+} + H_{\sigma^-}$, with $H_{\sigma^\pm} = V_\pm e^{\mp i\phi} + V_\pm^\dagger e^{\pm i\phi}$, $V_+ = \alpha^* |1'\rangle\langle 2|$ and $V_- = \alpha |2'\rangle\langle 1|$ with $\alpha = -(d_{12}\varepsilon_{02}/2) e^{-i\omega t}$ and $d_{12} = \langle 1|\vec{d}\vec{e}_z|2\rangle$, where $\vec{d}$ is the transition dipole moment. The total wave function of the atomic system is given to the first order of perturbation with respect to the σ pulse by [13]:

$$|\psi(t)\rangle = |\psi^{(0)}(t)\rangle + e^{-i\phi}|\psi^{(\sigma+)}(t)\rangle + e^{i\phi}|\psi^{(\sigma^-)}(t)\rangle, \quad (1)$$

with

$$|\psi^{(0)}(t)\rangle = U(t,-\infty)|\psi(-\infty)\rangle, \quad (2a)$$

$$|\psi^{(\sigma^\pm)}(t)\rangle = -\frac{i}{\hbar}\int_{-\infty}^{t} U(t,t')V_\pm(t')|\psi^{(0)}(t')\rangle dt'. \quad (2b)$$

$U(t,t')$ is the time evolution operator under the strong π-polarized pulse whose nonvanishing elements are $U_{11} = U_{1'1'} = \cos(\theta_{t'}^t/2)$, $U_{22} = U_{2'2'} = \cos(\theta_{t'}^t/2) e^{-i\omega(t-t')}$, $U_{21} = -U_{2'1'} = i\sin(\theta_{t'}^t/2) e^{-i\omega t}$, $U_{12} = -U_{1'2'} = i\sin(\theta_{t'}^t/2) e^{i\omega t'}$ with $\theta_{t'}^t = \theta(t) - \theta(t')$, $\theta(t) = \frac{d\varepsilon_{01}}{\hbar} \times \int_{-\infty}^{t} f_1(0,t')dt'$ and $\theta_1 = \theta(t \to +\infty)$ is the pulse area on the transition $|1\rangle \to |2\rangle$.

It follows from Eqs. (1) and (2), that the time sequence of the interaction can be described as follows: the strong field turns on at $t = -\infty$ mixing the states $|1\rangle$ and $|2\rangle$ to intermediate time t', where the $\sigma_-(\sigma_+)$ couples $|1\rangle \to |2'\rangle$ ($|1'\rangle \to |2\rangle$), that are finally remixed by the strong field from intermediate time t' to the observation time t. Because of the σ_+ and σ_- components, the amplitude of transition between $|1'\rangle$ and $|2'\rangle$ can be seen as the superposition of the amplitudes of two quantum paths [Fig. 1(d)] involving photon absorption on the transition $|1\rangle \to |2'\rangle$ and photon stimulated emission on the transition $|2\rangle \to |1'\rangle$. This is an original situation where interference effects take place between two superposition states $(\{|1\rangle, |2\rangle\} \to \{|1'\rangle, |2'\rangle\}$, and are observable only because the strong π-polarized pulse mixes both the initial states and the final states. An important result here is that the population in these states exhibits interferences with 2ϕ oscillations since the phase factor associated with absorption and emission paths are complex conjugate. When the intensity of the π-polarized pulse is sufficiently low so that a perturbative picture with respect to the π-polarized pulse holds, these two paths can be also interpreted in terms of Raman transitions. For instance, the amplitude in state $|1'\rangle$ results from the superposition of the Raman path $|1\rangle \xrightarrow{\pi} |2\rangle \xrightarrow{\sigma^+} |1'\rangle$ and $|1\rangle \xrightarrow{\sigma^-} |2\rangle \xrightarrow{\pi} |1'\rangle$. The amplitude in state $|2'\rangle$ results from the superposition of one-photon excitation path $|1\rangle \xrightarrow{\sigma^-} |2'\rangle$ and a three photon double Raman excitation path $|1\rangle \xrightarrow{\pi} |2\rangle \xrightarrow{\sigma^+} |1'\rangle \xrightarrow{\pi} |2'\rangle$. However, in this last case the interference effects would not be observable since the amplitude of the two paths would be incommensurate. A strong intensity for the π-polarized pulse is then required to balance the two paths. The perturbative interpretation thus breaks down. The mixing induced by the strong field necessitates the two-path picture as the relevant interpretation.

An alternative description of the interaction is the adiabatic representation where the atomic system is dressed by the π-polarized pulse. This description gives a complementary insight into the interaction and makes clear the significance of the ϕ dependence for the transmitted σ pulse. The adiabatic states are defined by $|\pm\rangle = (e^{-i\omega t}|2\rangle \mp |1\rangle)/\sqrt{2}$ and $|\pm'\rangle = (|1'\rangle \pm e^{-i\omega t}|2'\rangle)/\sqrt{2}$. In this representation, the coupling due to the σ pulse [Fig. 1(b)] leads to the following nonvanishing coupling elements:

$$\langle +|H_\sigma|+'\rangle = -\langle -|H_\sigma|-'\rangle = i\hbar\chi_2(t)\sin\phi, \quad (3a)$$

$$\langle +|H_\sigma|-'\rangle = -\langle -|H_\sigma|+'\rangle = -\hbar\chi_2(t)\cos\phi, \quad (3b)$$

with $\chi_2(t) = d\varepsilon_{02} f_2(0,t)/\hbar$. The key point in this description is the distinction that appears between the parallel and antiparallel adiabatic states, highlighting the ϕ dependence [Fig. 1(b)]. The parallel states are coupled through the imaginary part of the σ pulse field—proportional to $\sin\phi$—whereas the antiparallel states are coupled through the real part—proportional to $\cos\phi$. The phase variation from 0 to $\pi/2$ leads to a progressive evolution from nonresonant (antiparallel) coupling to resonant (parallel) coupling, performing a real control of light shifts effects. We shall see now how these light shifts and interference effects influence the transmitted σ pulse energy that is experimentally detected. As it propagates through the medium, the σ pulse obeys the paraxial equation of propagation [14] ($\partial_Y \equiv \partial/\partial_Y$):

$$\partial_Y[f_2(Y,t)e^{i\phi}] = -i(e_{\text{disp}}/\theta_2)\rho^{(\sigma)}(Y,t), \quad (4)$$

with $\rho^{(\sigma)} = \rho_{2'1} + \rho_{21'}$, $\rho_{ij} = \langle i|\rho|j\rangle$ and $\rho = |\psi\rangle\langle\psi|$, $Y = y/L$, with L the length of the medium, N is the atomic density in the sample, $\theta_2 = d\varepsilon_{02}\tau_0/\hbar$ is the σ pulse area, $e_{\rm disp} = \frac{NLd^2\omega\tau_0}{2\varepsilon_0 c\hbar}$ parameter that characterizes the severity of dispersion effects. We assume next that the π-polarized pulse is strong enough so that it is only slightly distorted during propagation (in our numerical simulations we take into account implicitly the propagation effects for the strong pulse). At the lowest order of the σ pulse and from Eqs. (1) and (2), we can recast the coherence $\rho^{(\sigma)}(t)$ for $Y = 0$ as

$$\rho^{(\sigma)}(t) = \rho_{\|}(t)\sin\phi + \rho_{\not\|}(t)\cos\phi, \tag{5}$$

with $\rho_{\|}(t) = \cos\theta(t)\int_{-\infty}^{t}\chi_2(t')dt'$ and $\rho_{\not\|}(t) = -i\int_{-\infty}^{t}\cos\theta(t')\chi_2(t')dt'$. The first term (second term) on the right in (5) represents the contribution to the coherence of transitions between parallel $\|$ (antiparallel $\not\|$) states. For the values of the density for which $e_{\rm disp} < 1$, and by restricting time t to $t \leq \tau_0/e_{\rm disp}$, the transmitted intensity $I_2(t) = |\varepsilon_{02}f_2(Y = 1, t)|^2$ can then be rewritten as

$$I_2(t) \simeq |\varepsilon_{02}f_2(Y = 0, t)e^{i\phi} + \beta(\rho_{\|}(t)\sin\phi + \rho_{\not\|}(t)\cos\phi)|^2 \tag{6}$$

with $\beta = -i\varepsilon_{02}e_{\rm disp}/\theta_2$. For $\phi = 0$, the contribution of parallel states vanishes and the contribution of the antiparallel states decreases as $\theta(t) > 2\pi$. So for large intensities of the π-polarized pulse we get $I_2(t) \simeq |\varepsilon_{02}|^2|f_2(0, t)|^2$: the σ pulse is transmitted without any modification of either its shape or energy. The combined effects of absorption and emission processes are canceled for $\phi = 0$. When $\phi = \pi/2$, the contribution between antiparallel states vanishes and $I_2(t) \simeq |\varepsilon_{02}|^2|f_2(0, t) - e_{\rm disp}\cos\theta(t)\int_{-\infty}^{t} f_2(0, t')dt'|^2$. During the action of the strong π-polarized pulse, the dynamics alternates between regimes where amplification $[\cos\theta(t) < 0]$ or absorption $[\cos\theta(t) > 0]$ of the σ pulse occur. Indeed, the dependence on the strong pulse is related to the induced Rabi oscillations on both transitions $1 \leftrightarrow 2$ and $1' \leftrightarrow 2'$. A reinforcement of the emission process occurs as long as inversion of population between levels $S_{1/2}$ and $P_{1/2}$ takes place. In the opposite case absorption dominates the dynamics. However, this absorption is only transient since at the end of the π-polarized pulse, the energy stored in the excited state is restored back coherently to the exciting field by free induction decay. No permanent deposition of energy into the medium is expected since the relaxation times are very large. The temporal profile of the transmitted σ pulse exhibits thus during the action of the π pulse a modulation pattern that maps out the Rabi oscillations and on the long time scale oscillations corresponding to the usual ringing behavior of free induction decay radiation (characteristic time $\tau_0/e_{\rm disp}$) [15].

Experimental observation of these interferences has been successfully demonstrated. A regenerative amplifier pumped by a Ti:sapphire laser delivers linearly polarized laser pulses with $\tau_0 = 90$ fs. 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 $\lambda/2$ wave plate combined with a polarizer rotates the polarization by 90° and allows an eventual modification of the energy of the pulse. The beam emerging from this arm constitutes the strong π-polarized pulse whereas the σ pulse propagates in the other arm. The pulse energy and waist are 45 μJ and 1.1 mm for the π-polarized pulse and 0.14 μJ and 0.4 mm for the σ pulse. The estimated pulse areas at the cell entrance are then $\theta_1 \simeq 1.1\pi$, and $\theta_2 \simeq 0.2\pi$, and the Rayleigh lengths are 570 and 60 cm, respectively, larger than the length of the cell (12 cm). The temperature into the cell was set to $T \simeq 130\,°$C for which $e_{\rm disp} \simeq 0.11$. At the exit, the two pulses are separated by a polarizer and the transmitted energy of the σ pulse is measured with a photodiode. Experimental results are displayed in Fig. 2. The energy of the σ pulse is modulated with a period of about 1.36 fs, close to the half of the optical period (2.65 fs at $\lambda = 794.76$ nm) demonstrating the interference effect presented in this Letter. It is important to emphasize that the interference process presented here strongly contrasts with that in temporal-Ramsey fringes or wave-packet interferometry [16,17], where a sequence of two time-delayed pulses excites an atomic transition leading to a modulation of the transferred population at the transition frequency. This transition frequency coincides with laser frequency except in the situation of multiphoton transition where it is equal to a multiple of the laser frequency [17], and modulation results from the interference between two time-delayed photon-absorption paths. In our situation, the interference process results from the competition between one-photon-absorption path and one-photon-emission path from the σ pulse. Although the process is linear with respect to the σ pulse, the modulation frequency is twice the transition frequency. Finally, these interference effects take place although the two fields have orthogonal polarizations and do not interfere. These properties reveal the original nature of these interferences.

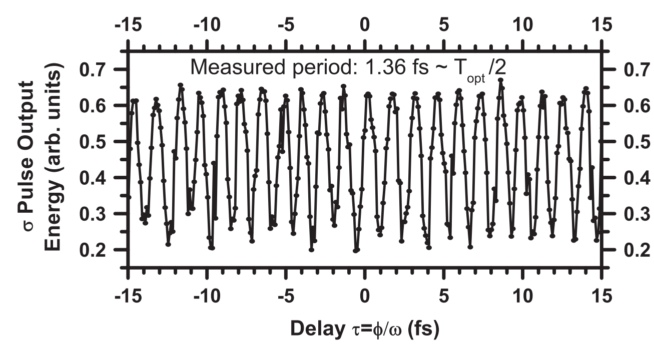

FIG. 2. Output energy of the σ pulse. Oscillations occur although the weak and the strong pulse are orthogonally polarized. The measured period is half the optical period. See text for experimental parameters.

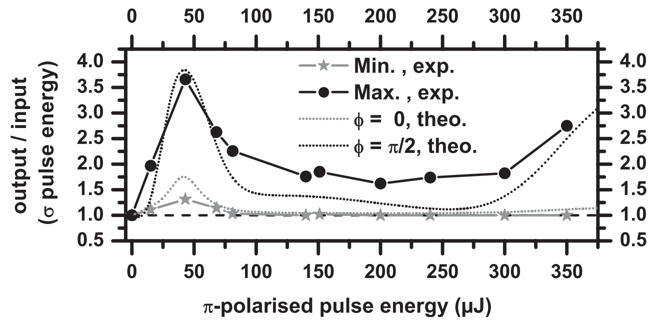

FIG. 3. Dependence of the normalized output energy of the σ pulse with the energy of the strong pulse for delay giving the maximum and minimum value, and compared with the theoretical curves obtained for $\phi = 0$ and $\phi = \pi/2$, respectively. The traces show with one and half Rabi oscillation (see text for parameters).

In Fig. 3 we represent the dependence versus the π-polarized pulse energy and for two relative delays corresponding to the maximum and minimum of the transmitted σ pulse energy, corresponding to $\phi = 0$ and $\phi = \pi/2$, respectively. The theoretical curves were obtained by solving numerically the Maxwell-Bloch equations. We take into account the influence of $P_{3/2}$, the $D_{5/2}$ and $D_{7/2}$ terms, the continuum, and the residual chirp of the pulses (1500 fs^2). The energy of the π-polarized pulse has been varied up to a limit of about 350 μJ. Good agreement is obtained between the experimental and theoretical curves. At fixed energy there is a significant gain modification between the case $\phi = 0$ and $\phi = \pi/2$. For $\phi = 0$, the σ pulse connects only antiparallel states and this interaction becomes nonresonant as the energy of the strong π-polarized pulse increases. Thus, the transmitted energy is almost unaffected by the propagation through the medium. For $\phi = \pi/2$, the transmitted energy oscillates with the π-polarized pulse energy, a signature of the Rabi oscillations that take place in the system. A peak of amplification (factor $\sim$4) is observed for $\theta_1 \simeq \pi$ and corresponds to a situation where a maximum of inversion of population is realized at the end of the strong pulse. Note that the global loss for the σ pulse is always negligible even when $\theta_1 \leq \pi/2$ for which there is no inversion of population during the whole process: the energy deposited into the medium is always restored coherently to the field by free induction decay.

In conclusion, we have shown in a double two-level system driven by a strong linearly polarized pulse, that light-shift effects can be controlled for a weak propagating pulse by varying the relative phase between the two pulses. An interesting feature here is the possibility to use either of two independent parameters (the relative phase and the strong pulse intensity), to switch off/on the interaction for the weak pulse, and to vary the medium gain. This contrasts with EIT experiments [4], where the strong field can only switch the interaction from absorption to transparency with no possibility to modify the gain. The results presented here may open the way to interesting extensions in strong field experiments with the introduction of an additional and efficient parameter of control. They could also be applied to improve gate switching in ultrafast optical processing.

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