Accumulative effects in temporal coherent control

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This paper analyzes the result of a temporal coherent control experiment using trains of ultrashort laser pulses in the situation where the laser repetition period is smaller than the relaxation times of the system. We focus on the accumulative effects present in sequential two-photon absorption. It is shown that stimulated emission due to the accumulation of population in the excited state give an important contribution to the coherently controlled process. Excellent agreement is obtained in the comparison between experiments in rubidium vapor and theory, where resonant pulse propagation is also taken into account.

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Temporal coherent control denotes the set of techniques that, in the optical domain, employ combinations of ultrashort laser pulses with a variable relative delay to control the outcome of a given quantum process. It has been applied to a wide range of systems, such as atoms [1,2], molecules [3–5], and excitons in semiconductor quantum wells [6]. The motivation varies from one system to another. In molecules, for example, there is an interest in the detailed control of chemical reactions [7], while in semiconductors, one goal is the development of optical switches for ultrafast optoelectronics [6,8]. The simplest systems, such as alkali atoms and diatomic molecules, play the role of prototypes: for these, it is possible to develop theories that are easier to compare with the experimental data.

The most conventional configuration for temporal-coherent control employs pairs of laser pulses of equal shapes to both excite and probe the sample. In such scheme, the observation of interferometric beat signals (periods of a few femtoseconds) in atoms and diatomic molecules have received great attention, since they are directly connected to quantum interferences between different excitation pathways [1,2,4,5,9]. A beautiful and direct extension to semiconductor quantum wells of such interferometric experiments can be found in Ref. [10]. For simple molecular systems [4,5], there are also a great number of measurements concerning the dynamics of vibrational wave packets [3–5]. Most of the studies with atoms and diatomic molecules were performed at very low densities to avoid propagation effects on the laser pulses. An experimental investigation in atomic rubidium varying the atomic density is reported in Ref. [11], where propagation effects lead to the formation of 0π pulses [12,13] and produce density-dependent beating signals with periods of a few picoseconds.

In general, the experiments performed with atoms, however, leave out an important issue: the atomic levels may have lifetimes of more than 10 ns, while the repetition rate of the most commonly used lasers (especially, Ti:sapphire) is of about 100 MHz, which gives a time interval of 10 ns between two consecutive pulse pairs. These two points considered together indicate that some accumulative effect must be present in such systems. Accumulative effects with cw mode-locked lasers were first observed in photon-echo experiments [14,15] and are an important issue for coherent control in semiconductor quantum structures to be used as high-speed optical switches [8,16]. In these systems, the incoherent accumulation of population in the excited state can prevent an efficient turnoff of the switch.

In this work, we discuss accumulative effects in the temporal-coherent control of a three-level atom in a cascade configuration [Fig. 1(a)]. We consider a sequence of pulse pairs separated by the laser repetition period T_R, in which the two pulses in the pair have a temporal relative delay of τ [Fig. 1(b)]. Both τ and the pulse duration are assumed much smaller than T_R or any relaxation time of the system. A theory in the weak-field regime is developed for the case where population accumulates only in level 3: the one with greatest lifetime. It is shown that, besides an increase in the population of the third level, such accumulation leads to interferometric terms whose average over an optical period does not vanish. These terms contribute even for averaged low-resolution scans of the temporal delay line, originating a beating at the frequency difference of the two transitions. Finally, the theory is compared with experimental results obtained in Rb vapor and good agreement is obtained for both high- and low-resolution scans of τ.

Our starting point is the Liouville equation for the density-matrix elements of a three-level system

$$\frac{d\rho_{kl}}{dt} = -\frac{i}{\hbar}\langle k|[H,\rho]|l\rangle - \frac{1}{T_{kl}}\rho_{kl}, \tag{1}$$

where H is the system Hamiltonian in the presence of the electric field $E(t)$ of a laser pulse, k and l denote states k and l, respectively, and T_{kl} is the relaxation time of the kl density matrix element. From Eq. (1), we can obtain the population $\rho_{33}(t)$ of level 3 for times much longer than the pulse duration, but shorter than every T_{kl} or T_R. Since we are only interested in an accumulation of the third-level population,

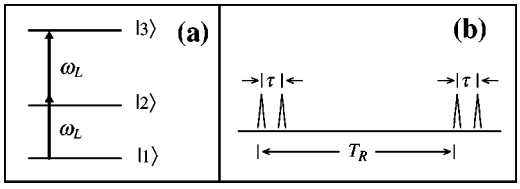

FIG. 1. (a) Cascade configuration for a three-level atom and (b) temporal diagram for a sequence of pulse pairs.

we consider that, from all initial density-matrix elements $\rho_{kl}(t=0)=\rho_{kl}^0$, only the populations in the initial and final states are nonzero: $\rho_{11}^0,\rho_{33}^0\neq 0$. With this in mind and since $\rho_{11}^0=1-\rho_{33}^0$, we obtain the following expression for ρ_{33}, in the lowest order of the field:

$$\rho_{33}(t)=e^{-t/T_{33}}\rho_{33}^0[1-S]+e^{-t/T_{33}}C, \tag{2}$$

where C is a second-order term giving the usual two-photon feeding of the third level by the population in the first level and S is a first-order term giving the reduction of the initial population ρ_{33}^0 caused by stimulated emission. The factor $e^{-t/T_{33}}$ gives the incoherent decay of the population with the lifetime T_{33} of level 3.

For temporal coherent control, the laser pulses are divided into pairs of pulses of equal shape, separated by a temporal delay τ, that is, $E(t)=E_1(t)+E_1(t-\tau)$. In this case, the factors S and C become functions of τ and are given by

$$S(\tau)=h_1[1+\cos(\omega_{32}\tau)], \tag{3}$$

and

$$C(\tau)=|F(0)+e^{i\omega_{21}\tau}F(-\tau)+e^{i\omega_{32}\tau}F(\tau)+e^{i\omega_{31}\tau}F(0)|^2, \tag{4}$$

with

$$h_1=2\left|\frac{\mu_{23}}{\hbar}\int_{-\infty}^{\infty}dt'e^{i\omega_{32}t'}E_1(t')\right|^2, \tag{5}$$

$$F(\tau)=\frac{\mu_{12}\mu_{23}}{\hbar^2}\int_{-\infty}^{\infty}dt'\int_{-\infty}^{t'+\tau}dt'' \times e^{i\omega_{32}t'}e^{i\omega_{21}t''}E_1(t')E_1(t''), \tag{6}$$

where μ_{12} and μ_{23} are the dipole moments of the first and second transition, respectively, and ω_{ij} is the transition frequency from state j to state i. Note that, as expected for a stimulated emission term, the integral in h_1 is simply the pulse spectrum at the frequency of the second transition.

The function $C(\tau)$ constitutes one of the basic results for the temporal coherent control of two-photon transitions and a full description of all its terms can be found, for example, in Ref. [5]. The four terms represent the different paths that an atom initially in the ground state can take to reach the third state. When the square modulus is taken, the resulting 16 terms give the probability for each one of these processes plus the quantum interferences between them. The existence of such interferences is the whole point of the coherent control, since it allows for large modulations of the population by varying the relative delay τ. The separate probabilities of the different pathways constitute a background term, that depends on τ only through the slowly varying function $F(\tau)$. From Eq. (4), we see that the interferences occur at frequencies ω_{21}, ω_{32}, ω_{31}, and $\omega_{32}-\omega_{21}$. The first three of these result in the interferometric beatings. The contribution with the frequency difference is not interferometric. This term, however, contributes only in a region near $\tau=0$. For τ values much greater than the pulse width at half maximum only the interferometric terms plus the background remain.

Now consider ρ_{33}^j as being the initial population of level 3 preceding the jth pulse pair. From Eq. (2), we see that ρ_{33}^{j+1} is equal to $\rho_{33}(T_R)$ when $\rho_{33}^0=\rho_{33}^j$. After a large number of pulse pairs, the population of level 3 must reach a stationary value. In this situation, a self-consistency condition applies: $\rho_{33}^{j+1}=\rho_{33}^j=\rho_{33}^f$. From Eq. (2) this results in

$$\rho_{33}^f=\frac{\gamma_{33}C(\tau)}{1+\gamma_{33}S(\tau)}, \tag{7}$$

with $\gamma_{33}=e^{-T_R/T_{33}}/(1-e^{-T_R/T_{33}})$, that is an amplification factor. When $T_{33}\gg T_R$, for example, γ_{33} is simply T_{33}/T_R, the number of pulse pairs that arrive during the relaxation time of level 3.

The simplest limit of Eq. (7) occurs when $\gamma_{33}S\ll 1$. In this case, $\rho_{33}^f\approx\gamma_{33}C(\tau)$ and the accumulative effect results in a multiplicative constant in front of the commonly known expression for the population. This is the situation, for example, when we have a purely two-photon transition, so that $S=0$, as in the studies with Cs atoms [1,2]. All interferometric terms in $C(\tau)$ disappear for low-resolution scans of τ when averaging over optical periods [5]. As these are the only beat signals that survive for τ much longer than the pulse width at half maximum, we should not observe any beating in this situation.

When $\gamma_{33}S\gtrsim 1$, on the other hand, the picture is quite different. The denominator in Eq. (7) leads to expressions for the interferometric terms whose average over an optical period does not vanish: it gives rise to a contribution with frequency $\omega_{32}-\omega_{21}$ as long as the interferometric terms are present. These terms survive for much longer temporal separations than the pulse width. Thus, low-resolution scans of τ with averaging over optical periods can distinguish this situation from the one described in the last paragraph. This behavior has already been observed and reported [11], with a beating in the difference frequency clearly present for τ values greater than 40 ps, in a situation where the pulse widths at half maximum were smaller than 1 ps. Reference [11], however, lacks a quantitative theoretical explanation of the observed features, which is now provided.

In our experiments, a vapor of Rb atoms is excited by pairs of laser pulses tuned to $\lambda_L=778$ nm ($\Delta\lambda_L\approx 8$ nm), which corresponds to the sequential $5S$-$5P_{3/2}$-$5D$ transition of Rb. For comparison with the theory described above, levels $5S$, $5P_{3/2}$, and $5D$ correspond to levels 1, 2, and 3, respectively. The excitation source is a Ti:sapphire laser generating pulses of about 100 fs duration with a repetition rate of 76 MHz, which corresponds to $T_R=13$ ns. Since we have $T_{33}=241$ ns for the $5D$ state of Rb, the amplification factor is $\gamma_{33}=18$.

The Rb vapor is enclosed in a 5-cm-long sealed cell, which can be heated to control the atomic density. Typically, we vary the atomic density from 10^{12} to 10^{14} atoms/cm^3. At these densities, propagation effects need to be taken into account and as shown in [11], we see the formation of 0π pulses in the medium [13]. To certify that we were really

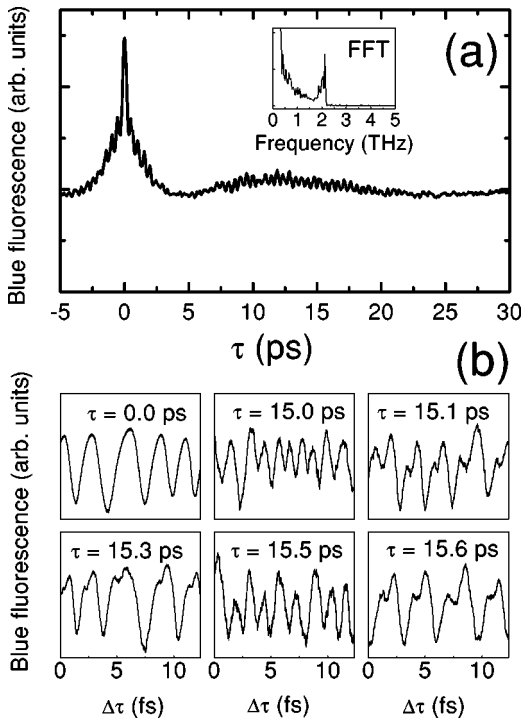

FIG. 2. Variation of the blue fluorescence with the relative temporal delay between the pulses in the pair. (a) Low-resolution scan. Inset shows the Fourier transform of the signal. (b) High-resolution scans around six different delays. The atomic density is 7×10^{12} atoms / cm^3.

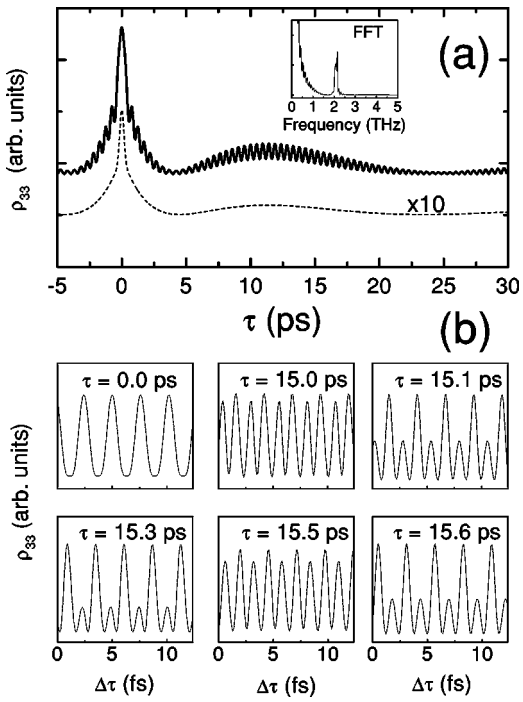

FIG. 3. The solid line is the variation of the population in the third level with temporal delay between pairs of pulses, as given by Eq. (7). The dashed line is the theoretical result in the absence of accumulation. (a) Low-resolution scan averaging over an optical period. Inset shows the Fourier transform of the solid line. (b) High-resolution scans around six different delays.

observing 0π pulses, we attenuated the laser intensity by an order of magnitude at the entrance of the cell, and noticed that the propagated pulse shapes measured by cross-correlation measurements remained unchanged. This observation also calls for a weak-field approximation in the theory. The 0π pulses are formed by the interaction of the laser pulses with the strong $5S$-$5P_{3/2}$ atomic transition. This results in the depletion of the ω_{21} spectral component of the laser pulse and in a negligible transfer of population from $5S$ to the $5P_{3/2}$ intermediate state. This effect and the faster decay time of the $5P_{3/2}$ state ($T_{22}=26.7$ ns) lead to a negligible accumulation of population in this level, and justify our approximation of accumulation only in the population of the third level.

The population in the $5D$ state relaxes to the $6P_{3/2}$ level and the blue fluorescence from this level is collected in a right angle geometry from the center of the cell, where the beam waist was measured to be 70 μm. The laser power in the cell is 100 mW and we used $\mu_{23}=4.24\times10^{-30}$ C m. For hyperbolic secant pulses at the entrance of the cell, these parameter values result in $h_1\sim0.05$ and $\gamma_{33}h_1\sim0.9$. Thus, our experimental situation fits well the case in which $\gamma_{33}S\approx1$.

Typical experimental results are presented in Fig. 2, where the variation of the blue fluorescence with τ is plotted. In Fig. 2(a), it is shown a low-resolution scan of τ including the slowly varying background. It is clear the presence of a 473 fs beating that extends for tens of picoseconds [inset of Fig. 2(a)]. This is exactly the period corresponding to the frequency $\omega_{32}-\omega_{21}$. The slower oscillation forming an envelope for the 473 fs beat comes from pulse propagation and varies with the atomic density [11].

In Fig. 2(b), we see the results for high-resolution scans around six different delays. Note first, the difference between the scan around $\tau=0$ and all others. Around $\tau=0$, we see a pattern with just one oscillating frequency: ω_L, the central frequency of the laser pulses. Since we have a complete superposition of the two pulses at $\tau=0$, this behavior comes from the optical interference between them, and has a completely classical interpretation. For the other τ values, however, we note a beating at ω_L superposed with another at $2\omega_L$. The latter is a purely quantum contribution coming from the interference between different two-photon excitation pathways [1]. By varying the delay in intervals of 100 fs, we also notice a modification in the beating pattern that repeats itself with a period of about 500 fs. This is a consequence of the fact that, for time delays longer than the pulse width, the ω_L contribution is a combination of two close frequencies, ω_{32} and ω_{21}, which results in a pattern that is repeated with a 473 fs period.

The theoretical curves corresponding to this experimental situation are shown in Fig. 3. In Fig. 3(a), we have a low-resolution scan and in Fig. 3(b) six different high-resolution scans. Figure 3(b) is a direct application of Eq. (7), with $S(\tau)$ and $C(\tau)$ given by Eqs. (3) and (4), respectively. The solid line in Fig. 3(a) is obtained by averaging Eq. (7) over an optical period. The dashed line is an average over an optical period of Eq. (4), which gives the theoretical prediction for

the population with no accumulation [1,2,4]. The propagated field $E_1(t)$ is calculated using the well-known theory for 0π pulses [12]. The best fit to the experimental data was obtained for a pulse temporal width of $T_p = 140$ fs at the entrance of the cell, a strength of the electrical field such that $h_1 = 0.017$ and an absorption coefficient $\alpha_0 = 130$ cm^{-1}, which are compatible with the experimentally measured parameters.

For low-resolution scans, the comparison of the solid line in Fig. 3(a) with Fig. 2(a) shows good agreement. The theory predicts the correct beat frequency [inset of Fig. 3(a)], the amplitude relation between the varying signal and the background, and also the correct coupling between the 473 fs beating and the slow envelope due to pulse propagation. The only discrepancy that we could distinguish is in the width of the $\tau = 0$ peak, which could be due to a chirp of the laser pulses from elements outside the vapor cell. For the high-resolution scans, we obtain qualitative agreement between theory [Fig. 2(b)], and experiment [Fig. 3(b)]. The difference between the interferometric signal around $\tau = 0$ and that for τ much longer than the pulse widths is correctly predicted, with the presence of a $2\omega_L$ frequency component in this last case. It has also correctly predicted the variation of the signal for steps of 100 fs in the time delay. These characteristics of the interferometric signals are not dependent of any accumulation of population and can be inferred from the $C(\tau)$ function only, they are emphasized here just to show the consistency of our measurements and theory with previous works [1,5]. The low-resolution theoretical curve, on the other hand, has a shape that is dependent on the accumulation of population in level 3, as can be inferred from the comparison between solid and dashed lines in Fig. 3(a). Its accordance with the experimental observation, thus, confirms the adequacy of our treatment. Comparisons for others densities of the atomic vapor also show good agreement with the experimental results.

In summary, we have developed a theory for the temporal-coherent control of three-level systems that takes into account accumulation of population in its highest level caused by a train of laser pulses with interpulse separations much smaller than the relaxation time of the level. This theory predicts that, in a situation where stimulated emission in the second transition is relevant, the interferometric terms can still contribute to low-resolution scans of the delay time averaged over optical periods. This indicates the possibility of efficient temporal coherent control with much simpler experimental schemes. We report experiments in such situation and compare their results with the theory, obtaining very good agreement.

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