Observation of Coherent Transients in Ultrashort Chirped Excitation of an Undamped Two-Level System

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The effects of coherent excitation of a two-level system with a linearly chirped pulse are studied theoretically and experimentally [in Rb $(5s - 5p)$] in the low field regime. The coherent transients are measured directly on the excited state population on an ultrashort time scale. A sharp step corresponds to the passage through resonance. It is followed by oscillations resulting from interferences between off-resonant and resonant contributions. We finally show the equivalence between this experiment and Fresnel diffraction by a sharp edge.

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Coherent excitation of atomic transitions is a general phenomenon occurring whenever electromagnetic radiation interacts with atoms. This interaction may result in various kinds of processes, for example, Rabi oscillations, free induction decay, adiabatic population transfer, and coherent transients (CT) [1]. By simply varying the temporal shape of the pulses, a great variety of systems can be manipulated, such as spins in nuclear magnetic resonance [2] or atomic and molecular systems in coherent control schemes [3]. We consider here the simplest case of linearly chirped pulses where the "instantaneous frequency" drifts in time.

Depending on the frequency sweep and on the intensity, adiabatic following can be observed with a significant population transfer. For example, total population transfer has been achieved via multilevel ladder climbing [4–9] and stimulated Raman adiabatic passage [10].

On the contrary, if the adiabaticity criterion is not fulfilled, the final population depends crucially on the pulse integral and CT dominate the interaction. This Letter presents theoretical and experimental studies of these coherent transients. Although many works have dealt with the transfer efficiency into the final state, we report here the first direct observation of these transients on a subpicosecond time scale. The CT result in oscillating population. This is a signature of interferences between resonant and nonresonant excitation. A detailed understanding of this behavior is crucial to analyze pump-probe experiments in which dynamics takes place on the same time scale as laser interaction. Because of the difficulty to control properly the laser pulses, the effects of CT are most often ignored [11].

Similarities can be found with optical CT observed in the experiments of free induction decay or photon echoes which provide relaxation rates [12–14]. In these experiments, they observed on the transmitted optical signal, a beat between initially induced polarization of the medium at resonance and a near-resonant laser. In the particular case of a single chirped optical pulse, the beat frequency increases linearly after passage through resonance, following the increase of resonance mismatch [15]. Contrary to these previous studies, the present report involves CT directly observed on the population before relaxation becomes significant. The beats result here from interferences between the resonant and nonresonant excitation paths. The passage through resonance does not lead to total inversion [16]. Thus nonresonant excitation can lead to population transfer after resonance. However, these extra contributions have a phase which varies rapidly with respect to the phase of the dominant contribution, leading to population oscillations.

In this Letter, we describe theoretically the temporal evolution of the excited state population induced by chirped pulse excitation in the low field regime. The CT oscillating pattern is experimentally studied on the Rb $(5s \rightarrow 5p)$ transition. The $5p$ population is probed "in real time" by a second excitation induced by an ultrashort pulse overlapping partially with the excitation pulse. These results illustrate the relative importance of the various stages of the interaction. Most of the population transfer occurs at resonance. The small fraction of excited state amplitude transferred after resonance leads to strong interferences, whereas interaction before resonance results in negligible effects. In order to point out the different stages of the interaction, two cases are investigated, the first being when resonance is reached at the middle of the interaction (on-resonance case), and second when the resonance is reached earlier (off-resonance case).

A quantum system with three states $|g\rangle$, $|e\rangle$, and $|f\rangle$, of increasing energies $\hbar\omega_g$, $\hbar\omega_e$, and $\hbar\omega_f$, respectively, interacts with two ultrashort laser pulses $E_1(t)$ and $E_2(t)$. These laser pulses have carrier frequencies ω_1 and ω_2, each one close to resonance with an atomic transition: $\omega_{eg} = \omega_e - \omega_g \simeq \omega_1$ and $\omega_{fe} = \omega_f - \omega_e \simeq \omega_2$. Moreover, these transitions are sufficiently separated compared to the laser linewidths ($|\omega_{fe} - \omega_{eg}| \gg \Delta\omega_1, \Delta\omega_2$) so that each laser can be assumed to interact with one atomic transition only.

The first laser pulse is obtained by frequency chirping a Fourier transform limited laser pulse of duration $\tau_1 = \frac{4\ln 2}{\Delta\omega_1}$. The second one is kept as short as possible in order to probe the transient population. Keeping only the resonant component (within the rotating wave approximation), the electric fields of the pulses are given by (for $n = 1, 2$) $E_n(t) = E_n(0)\exp(-\Gamma_n t^2)\exp(-i\omega_n t)$ where $\Gamma_n = [(8\ln 2/\Delta\omega_n^2) - 2i\phi_n'']^{-1}$ and the new pulse duration is $\tau_{nc} = \tau_n\sqrt{1 + [(4\ln 2\phi_n'')^2/\tau_n^4]}$. Here $\phi_n'' = d^2\phi_n/d\omega^2$ is the quadratic dispersion responsible for the group velocity dispersion induced in the pulse by the chirping process. In this Letter, we concentrate on the case where the pump pulse is strongly chirped, so that $\tau_{1c} \simeq 2\phi_1''/\tau_1$. Moreover, to measure the "instantaneous" excited state population (in state $|e\rangle$), the probe pulse is assumed to be nearly Fourier transform limited ($\phi_2'' \simeq 0$) and of short duration compared to the chirped pump pulse duration ($\tau_2 \ll \tau_{1c}$). This will ensure that the final state population (in state $|f\rangle$) reproduces the $|e\rangle$ state population.

In the low field regime, analytic solutions of the Schrödinger equation are easily found within first order perturbation theory, with a probability amplitude of state $|e\rangle$ proportional to

$$a_e(t) \propto \int_{-\infty}^{t} \exp\left(-\frac{2\ln 2 t'^2}{\tau_{1c}^2}\right) \times \exp\left(-i\,\frac{(t' - t_0)^2 + t_0^2}{2\phi_1''}\right) dt', \qquad (1)$$

where $t_0 = (\omega_{eg} - \omega_1)\phi_1''$ is the time when the laser sweeping frequency goes through the atomic resonance.

In the stationary phase approximation, the main contribution to $a_e(t)$ arises from delays within the interval $[t_0 - \sqrt{\phi_1''}; t_0 + \sqrt{\phi_1''}]$. This corresponds to the interval during which the laser frequency is resonant with the atomic transition. Nonresonant transitions can also contribute to the excited state amplitude, but with a smaller efficiency. As time increases from t_0, new contributions are added to the amplitude $a_e(t)$. These contributions have a phase which increases more and more rapidly, leading alternatively to an increase and a decrease of the excited state amplitude.

In order to illustrate the coherent transients, an experiment has been performed in rubidium atoms. The excitation scheme is depicted in Fig. 1. The $[5s - 5p\ (^2P_{1/2})]$ transition (at 795 nm) is excited either with a Fourier transform limited pulse or with a chirped pulse. The laser bandwidth of ~10 nm limits the excitation of the other fine structure component $5p\ (^2P_{3/2})$ transition (at 780 nm). The transient excited state population is probed on the $(5p - ns, n'd)$ transitions with an ultrashort pulse (at 607 nm).

The laser system is based on a conventional Ti:sapphire laser with chirped pulse amplification (Spitfire Spectra Physics) producing 130 fs pulses around 795 nm at a 1 kHz repetition rate with energy of as much as 800 μJ. Half of the light is used as the pump pulse. The other half feeds a homemade noncollinear optical parametric amplifier [17] compressed with Brewster-cut prisms, which delivers pulses of a few micro-Joules, 30 fs (after compression), centered around 607 nm. The pump pulse is negatively chirped with a pair of gratings ($|\phi_1''| \gtrsim 2 \times 10^5$ fs^2), recombined with the probe pulse and sent into a sealed rubidium cell with fused silica Brewster-window ends (3 mm length). The cold finger of the cell is maintained at about 80 °C, corresponding to a pressure of 4×10^{-5} mbar. The pump-probe signal is detected by monitoring the fluorescence at 420 nm due to the radiative cascade $(ns, n'd) \rightarrow 6p \rightarrow 5s$ collected by a photomultiplier.

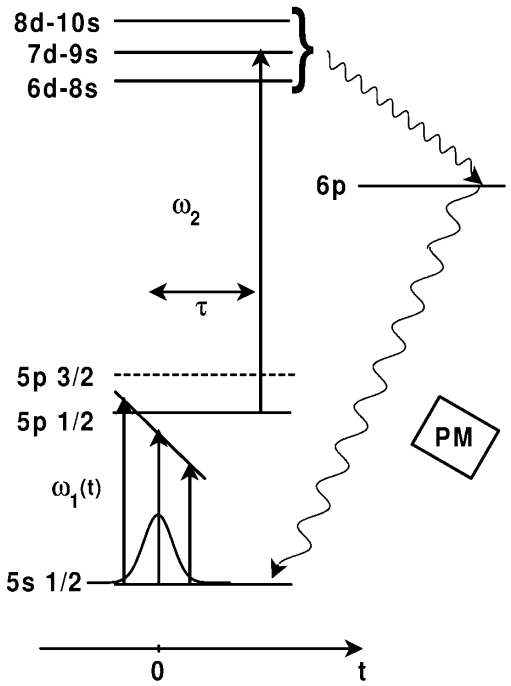

FIG. 1. Simplified energy-level diagram of rubidium. The pump pulse is chirped (and drawn here off-resonance).

Figures 2 and 3 present the experimental pump-probe signals. Measurements have been performed for resonant excitation ($\lambda_1 = 795$ nm on Fig. 2) and for a central wavelength detuned below resonance by roughly half the laser bandwidth ($\lambda_1 \simeq 801$ nm on Fig. 3). Each figure presents a fluorescence signal obtained with a chirp of $\phi_1'' = -4 \times 10^5$ fs^2 corresponding to a pulse duration of 11 ps. As a comparison, chirp-free data are displayed in the insets. In this latter case, the pump-probe signals show a sharp step, with a short rise time corresponding to the laser cross-correlation duration. On resonance, the plateau is slightly modulated by spin wave packet dynamics with a period of 140 fs. Indeed, off-resonance population of the $5p\ ^2P_{3/2}$ state is induced by the spectral tails of the pulse. The coherent superposition of the two excited states corresponds to a fine structure wave packet which oscillates [18,19]. These oscillations are well known and not related to the present study.

With a significant chirp, we observe large amplitude oscillations which are the signature of the coherent

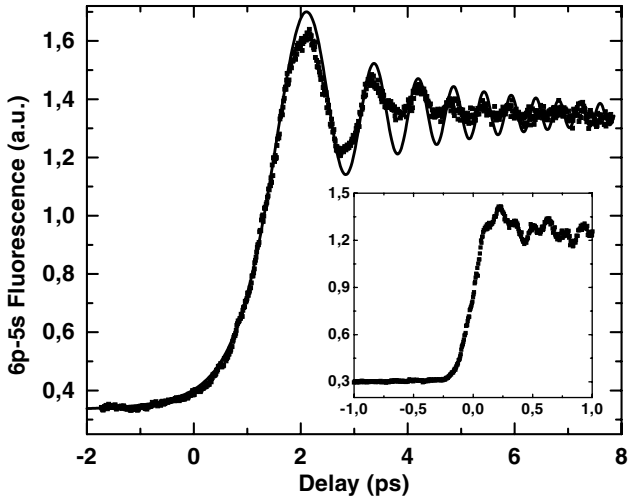

FIG. 2. Experimental results and numerical solution obtained on resonance ($\lambda_1 = 795$ nm) for a chirp of $\phi_1'' = -4 \times 10^5$ fs^2 and for a chirp-free pulse in the inset.

transients. The resonance experiment (Fig. 2) was performed in the perturbative regime (fluence of 2 μJ/cm^2). Because of the smaller efficiency of the off-resonance excitation ($\lambda = 801$ nm, Fig. 3), the fluence was increased to 110 μJ/cm^2, inducing some power effects. However, experiments performed for different fluences show qualitatively the same behavior.

In each case, the experimental curve is compared with the numerical solution of the Schrödinger equation projected on the $5s, 5p$ ($^2P_{1/2}, ^2P_{3/2}$), $6d$ up to $10s$ states. The comparison with the numerical resolution (where this time origin is well defined) has been used to adjust the relative time origins of the various experimental data. Indeed, in the case of long chirped pulse, the experimental determination of the time delay origin is a real challenge via standard setup based on frequency mixing. The CT scheme could provide an alternate experimental determination of both time origin and long linear chirp measurements. The only fitted parameter is the value of the chirp which differs by less than 2% from the estimates deduced from the geometry of the stretcher. An excellent agreement with the oscillation pattern is obtained. The contrast of the first periods is well reproduced. It drops more rapidly in the experiment. Several causes can be invoked: residual spatial chirps or small incoherent components of the pump pulse. After the sharp step, the oscillation frequency of the interferences increases linearly with time: $\omega_{eg} - \omega_1(t) = t/\phi_1''$. For a pulse frequency below resonance and a negative chirp, the passage through resonance occurs at the beginning of the pulse. Thus the major part of the pump spectral components contributes after the resonance so that many oscillations result as can be seen in Fig. 3. On the contrary, in the resonant case, only the second half of the pulse contributes to the interferences and fewer oscillations are observed.

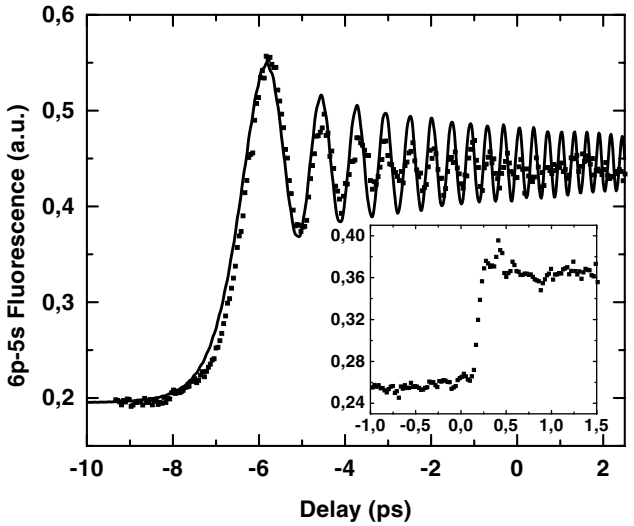

FIG. 3. Experimental results and numerical solution obtained below resonance ($\lambda_1 = 801$ nm) for a chirp of $\phi_1'' = -4 \times 10^5$ fs^2 and for a chirp-free pulse in the inset.

Another way to explain the CT phenomenon is to examine the behavior of $a_e(t)$ in the complex plane as displayed in the insets of Fig. 4 for $\phi_1'' = -8 \times 10^5$ fs^2. Two examples obtained via a numerical resolution of the Schrödinger equation are given. They correspond to the experimental data shown on the same figure. The probability amplitude follows a double spiral starting from $(0, 0)$. Three regions can be distinguished. The two spirals result from contributions before (I) and after (III) resonance. The intermediate region (II: time interval $[t_0 - \sqrt{\phi_1''}; t_0 + \sqrt{\phi_1''}]$) corresponds to the passage through resonance. It provides the main contribution to the population. This contribution is characterized by a sharp increase of $|a_e(t)|$, in an almost "straight" direction as expected from the stationary phase approximation applied to Eq. (1). The two spirals play different roles. The first one (I) winds around the origin with an increasing radius. The resulting probability increases thus slowly and regularly. After resonance (III), a second spiral winds around the asymptotic value leading to strong oscillations. For a central laser frequency equal to the resonance frequency, the two spirals have the same weight (Fig. 4a). Detuning of the laser frequency changes their relative weight (Fig. 4b). For negative chirp and negative detuning, the second spiral (after resonance) has a larger weight since resonance is reached during the first half of the pulse. This leads to a higher number of oscillations, as can be observed by comparing Figs. 2 and 3. Conversely, a negative chirp and positive detuning reduces the number of oscillations.

The general expression of the excited state amplitude [Eq. (1)] presents strong similarities with Fresnel diffraction of a Gaussian beam by a sharp edge. Effectively, the spirals displayed in Fig. 4 are similar to the well-known Cornu spirals. In Eq. (1) the variable t corresponds to the position of the sharp edge in the Gaussian beam. Without detuning, Eq. (1) reproduces the classical diffraction figure *on axis*. The effect of detuning can be understood by observing in a slant direction θ, with $\sin\theta \propto \Delta\omega$.

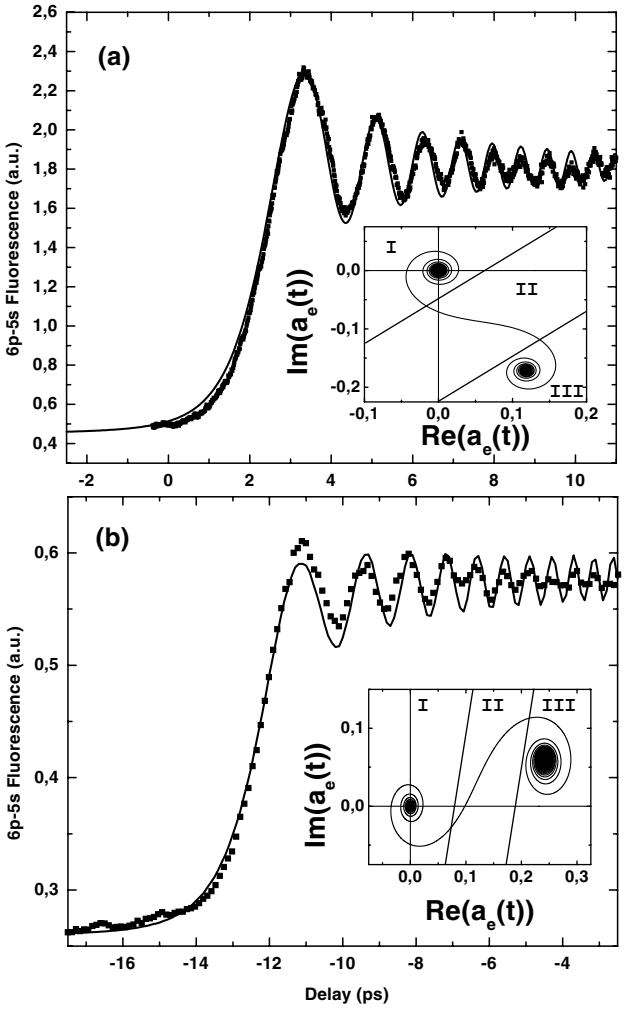

FIG. 4. Experimental results obtained (a) on resonance ($\lambda_1 = 795$ nm), and (b) below resonance ($\lambda_1 = 801$ nm) for a chirp of -8×10^5 fs^2 and the corresponding excited state amplitude (numerical resolution of the Schrödinger equation) drawn in the complex plane (insets).

Other analogies between time and space Fresnel diffraction have been reported by several groups [20,21]. One example deals with two-photon absorption (or frequency doubling) from a chirped pulse [20]. The spatial coordinate was associated with the laser frequency instead of the pump-probe delay in our present case. By inserting masks on the laser spectrum, the equivalent of a Fresnel zone lens was reproduced in the spectral domain. The frequency doubled pulse was thus spectrally focused. In our case, pulse shaping of the laser pump pulse should provide similar effects as a Fresnel lens. For instance, by slicing the temporal profile of the pump pulse, it should be possible to suppress the destructive interference contributions to $a_e(t)$ and increase significantly its asymptotic value.

We have presented in this Letter direct measurements of coherent transients observed on the excited state population at an ultrashort scale. The present experiments have been performed with a linear chirp. It provides an accurate measurement of the chirp. This scheme can be extended to nonlinear variations of the chirp which could thus be measured in a direct way. Moreover, in pump-probe experiments as well as in any experiment based on a combination of ultrashort pulses, such coherent transients should be taken carefully into account to analyze the data. Finally, based on this phenomenon, new pulse shaping schemes could be developed to improve the transfer efficiency, as demonstrated, for instance, in resonant multiphoton transition [22].

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