Realization of a Time-Domain Fresnel Lens with Coherent Control

Jérôme Degert,[1] Wendel Wohlleben,[2] Béatrice Chatel,[1] Marcus Motzkus,[2] and Bertrand Girard[1]

[1]*Laboratoire de Collisions, Agrégats, Réactivité (CNRS UMR 5589), IRSAMC, Université Paul Sabatier, 31062 Toulouse, France*

[2]*MPI für Quantenoptik, 85748 Garching, Germany*

(Received 30 April 2002; published 29 October 2002)

Perturbative chirped pulse excitation leads to oscillations of the excited state amplitude. These coherent transients are governed by interferences between resonant and off-resonant contributions. Control mechanisms in both frequency and time domain are used to modify these dynamics. First, by applying a phase step in the spectrum, we manipulate the phase of the oscillations. By direct analogy with Fresnel zone lenses, we then conceive highly phase-amplitude modulated pulse shapes that slice destructive interferences out of the excitation time structure and enhance the final population.

DOI: 10.1103/PhysRevLett.89.203003 PACS numbers: 32.80.Qk, 42.50.Md, 42.65.Re

Use of optimally shaped pulses to guide and control the temporal evolution of a system has been an active field of research in the past decade [1]. Recent experimental advances have led to successful control of molecular ionization and dissociation [2–4], atomic and molecular fluorescence [5–7], clusters reactivity [8], the shape of Rydberg wave functions [9] and excitations in semiconductors [10,11], but also of the energy flow in a biological system [12]. These papers were spurred by the technological breakthrough [13] for generating arbitrarily tailored pulses. However, in the absence of predesigned control mechanisms, only a closed loop scheme [14] may be employed to find efficient pulse shapes [3]: The outcome of many different shapes is fed back into an algorithm that iteratively optimizes the excitation shape without insight into the physical mechanism that is triggered by a particular shape. In contrast, the effect of shapes on small systems can be systematically studied within an open-loop scheme [6,15].

Our approach is based on this open-loop scheme. It consists of reaching a specific goal (manipulation of the temporal response of a system excited by a light pulse) without any experimental feedback. Physical analysis of the process allows us to predetermine the theoretical pulse shape which leads to the desired result. It is then implemented experimentally. To demonstrate this approach, we consider the simplest case of the excitation of a two-level system by a linearly chirped pulse. It leads to interferences between resonant and nonresonant excitation paths, which are the signature of coherent transients (CT) [16], first observed in the femtosecond scale in our previous work [17] and also studied in the saturated regime [18]. Recently, Silberberg *et al.* [19] showed how it is possible to enhance the transient population obtained in a two-level system with transform limited pulses by applying a π step on the spectral phase. In this work, we take advantage of the chirped pulses where the "instantaneous" frequency drifts in time, and combine it with amplitude and phase tailoring to control the full time evolution of the CT. For strongly chirped pulses, the time to frequency analogy can be used to understand the behavior of the CT. For instance, applying a spectral phase step at a given frequency results in a phase shift of the oscillations in the CT. Furthermore, we will demonstrate how to create a Fresnel lens in time domain: Analogous to focusing of light by partially obstructing the spatial profile, we slice the temporal structure of excitation pulses. CT oscillations are thus transformed into a climbing staircase pattern that decrease or increase the final transfer efficiency. Chirped pulses have already been used to create the equivalent of a Fresnel zone lens [20,21] in the spectral domain instead of the temporal domain in our case. A two-photon transition was, for instance, optimized by suppressing the frequencies leading to destructive interferences [20].

We recall first the origin of the coherent transients [17]. Consider the resonant interaction between an atomic system and a weak femtosecond laser pulse. First order time dependent perturbation theory predicts the temporal amplitude of the excited state to be [19]

$$a_e(t) = \frac{\mu_{eg}}{\hbar}\left\{\frac{1}{2}E(\omega_{eg})e^{i\phi(\omega_{eg})} + \frac{i}{2\pi}\wp\int_{-\infty}^{+\infty} d\omega\,\frac{e^{-i[(\omega-\omega_{eg})t-\phi(\omega)]}}{\omega-\omega_{eg}}E(\omega)\right\}, \tag{1}$$

where ω_{eg} is the transition frequency, μ_{eg} the dipole moment matrix element, $|g\rangle$ and $|e\rangle$ the ground and excited states, respectively, and $E(\omega)$ and $\phi(\omega)$ the amplitude and phase of the pump electric field spectrum. $\wp$ is the principal Cauchy value. The first term in Eq. (1) corresponds to the resonant contribution of the excited population, whereas the second term deals with the off-resonance contributions. CT result from interferences of these two terms. Naturally, most of the population transfer occurs at resonance. The small fraction of excited amplitude transferred after resonance leads to strong interferences, whereas interaction before resonance results in negligible effects [17]. With a significant chirp,

we observe large amplitude oscillations as depicted in Fig. 2 (below). In this work, we apply an additional phase or phase-amplitude shaping to dephase or modify these CT features.

Atomic rubidium is used as a benchmark system. The $5s$-$5p(2P_{1/2})$ transition (at 795 nm) is resonantly excited with a chirped shaped pulse [Fig. 1(a)]. The laser bandwidth of ~8 nm limits the excitation of the other fine structure component $2P_{3/2}$ (at 780 nm). The transient excited state population is probed "in real time" on the $(5p\text{-}ns, n'd)$ transitions with an ultrashort pulse (at 607 nm).

The laser system [Fig. 1(b)] is based on a conventional Ti:sapphire laser with chirped pulse amplification (Spitfire Spectra Physics) which supplies 1 mJ-130 fs-795 nm pulses. Half of the beam is used as the pump pulse. The remaining seeds a homemade noncollinear optical parametric amplifier [22] compressed with chirped mirrors, which delivers pulses of a few microjoules, 30 fs-FWHM pulse intensity, centered around 607 nm. The pump pulse is negatively chirped with a pair of gratings, shaped with a programmable pulse-shaping device, recombined with the probe pulse, and sent into a sealed rubidium cell. All the experiments are performed in the perturbative regime (fluence of 12 μJ/cm^2). 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 tube as a function of the pump-probe delay. The pulse-shaping device [23,24] is a $4f$ setup composed of one pair each of reflective gratings and cylindrical mirrors. Its active elements—a double stack 128 pixels liquid crystal mask—is installed in the common focal plane of both mirrors. The predominant part of the phase, which is quadratic, is introduced by the Treacy stretcher [$\phi'' = (d^2\phi/d\omega^2)_{\omega_L}$, where ω_L is the pump laser carrier frequency]. Here $|\phi''|$ ranges between 5×10^5 fs^2 and 10^6 fs^2, corresponding to pulse duration τ_c between 10 and 20 ps. The pulse-shaping apparatus allows us to add modulation to the quadratic phase and to filter simultaneously the amplitude of the frequencies. Keeping in mind that the final excited state population is proportional to the energy of the pulse at resonance, we use rubidium itself to identify precisely the pixel corresponding to resonance. This is achieved by switching off the transmission pixel per pixel, until the minimum asymptotic population is reached. The width of the resonance curve is approximately 1.5 pixel or 0.4 nm, limited by the diffraction spot size on the SLM.

The first experiment consists in producing CT in opposite phase with normal CT. Because of the time-frequency correspondence for a strongly chirped pulse, a π jump in the frequency domain for frequencies below the resonance translates almost exactly to a change of sign of the electric field after resonance. Thus, since the oscillations of the CT are due to interferences between on- and after-resonance contributions, this change of sign results in a π-phase shift of the oscillations. The π step occurs at a chosen frequency ω_{step}: Experimentally, we set the phases of the pixels of frequency $\omega_{\text{pixel}} < \omega_{\text{step}}$ to π and the others to zero. Because of the limited resolution of the shaper, the resulting π phase step function can be described by

$$\phi_{\text{step}}(\omega) = \frac{\pi}{2}\{1 - \tanh[\beta(\omega - \omega_{\text{step}})]\}, \qquad (2)$$

where the slope β produces a smooth transition of the π jump between adjacent pixels. Experimental results and numerical integration of Eq. (1) are displayed in Fig. 2. As can be seen, the contrast of the oscillations and also the pattern deformation due to the passage by the π jump are in perfect agreement. The comparison of the unshaped coherent transients with theory (where the time origin is well defined) has been used to adjust the relative time origins between experiment and theory, and the value of the chirp which differs by less than 4% from

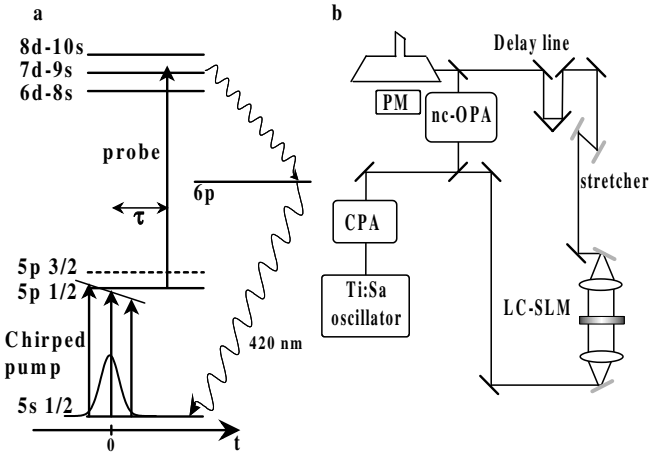

FIG. 1. (a) Excitation scheme of rubidium. τ is the pump-probe delay. (b) Experimental setup. The shaper is shown with lenses for clarity. CPA: regenerative amplifier; LC-SLM: liquid crystal spatial light modulator; PM: photomultiplier.

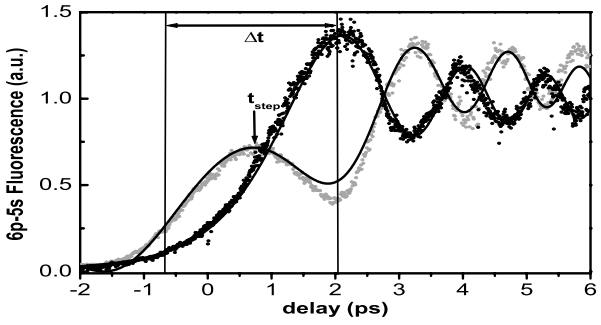

FIG. 2. Experimental (dotted) and numerical (solid) results: chirped pump pulse (black), and π-step shaped chirped pulse (gray). Here $\phi'' = -9.4 \times 10^5$ fs^2 corresponding to $\tau_c \simeq 21$ ps. Here $\lambda_{\text{step}} = 2\pi c/\omega_{\text{step}} = 795.23$ nm .

the estimates deduced from the geometry of the stretcher. Since the passage through resonance lasts approximately $\Delta t = 2\sqrt{2|\phi''|}$ ($\simeq 2.7$ ps), the transition domain from normal CT to inverted CT is approximately Δt broad centered on $t_{\text{step}} = (\omega_{\text{step}} - \omega_L)\phi''$ [see Fig. 2 (gray dots)] [25].

The second set of experiments reported in this work is based on the strong similarity between the expression of the excited state amplitude [17] and Fresnel diffraction of a Gaussian beam by a sharp edge. Equation (1) can be rewritten as a function of the temporal electric field,

$$a_e(t) \propto \int_{-\infty}^{+\infty} dt' \theta(t - t') \exp\left(-2 \ln 2 \, \frac{t'^2}{\tau_c^2}\right) \times \exp[-i(\delta\omega t' + \alpha t'^2)], \quad (3)$$

where $\theta(t)$ is the Heaviside step function, τ_c the chirped pulse duration, ω_L the laser carrier frequency, $\delta\omega = \omega_L - \omega_{eg}$ the detuning, and α the chirp rate. Here, t corresponds to the position of the sharp edge in Fresnel diffraction.

Thus, by slicing out sections in the pump pulse, the equivalent of a Fresnel zone lens can be reproduced in the time domain. So it should be possible to suppress the destructive (or constructive) contributions to $a_e(t)$ and increase (or decrease) significantly its asymptotic value by converting the oscillations into staircase patterns. Figure 3 shows a theoretical simulation of the climbing staircase pattern when a single cut is made in the time domain (inset of Fig. 3) during the first decreasing part of the CT. As expected, the final population is enhanced.

Slicing the pulse at a time scale of $\sim$100 fs is not directly possible in the time domain. As usual in fs pulse shaping, the required shape can be obtained by filtering the spectral profile. While decreasing the excitation energy [the area under spectra in Fig. 4(a)], the shaped temporal profile produces a redistribution of frequencies. In particular as shown on Fig. 4(a), the intensity (black line) at resonance (λ_{eg}) is increased compared to the original spectrum (black dashed line). Thus, the passive temporal filter of the inset of Fig. 3 corresponds to an active spectral filter.

We simulate this latter using a passive spectral filter, and applying an appropriate normalization factor by comparison of the shaped and unshaped pulse area. The required spectrum is obtained by Fourier transforming the shaped temporal profile (inset of Fig. 3). Its amplitude, represented in Fig. 4(a) (solid line), is highly structured. It was implemented experimentally by using the pulse shaper as a pure amplitude modulator. This leads to the spectrum (gray line) observed on Fig. 4(a). It has been smoothed by the finite resolution of the pulse shaper. However, the fastest oscillations are associated to the sharpness of the cutting, whereas the main features of physical interest (position and width of the temporal hole) are contained in the large peaks which are preserved even in the smoothed spectrum as our experiments have demonstrated. The corresponding phase is mainly quadratic (introduced by the stretcher as for unshaped CT) with some residual terms which have been neglected. Qualitatively, the agreement between the experimental result shown in Fig. 4(b) and the ideal result displayed in Fig. 3 is excellent. That includes the in-phase revival of oscillations after the suppressed interference, precisely as intended by the control goal.

To investigate the origin of the small quantitative discrepancies, we performed the inverse Fourier transform of the experimental spectrum together with the known quadratic phase. We obtain the temporal electric field of the experiment which is then used as an input field in the numerical resolution of the Schrödinger equation. Thus,

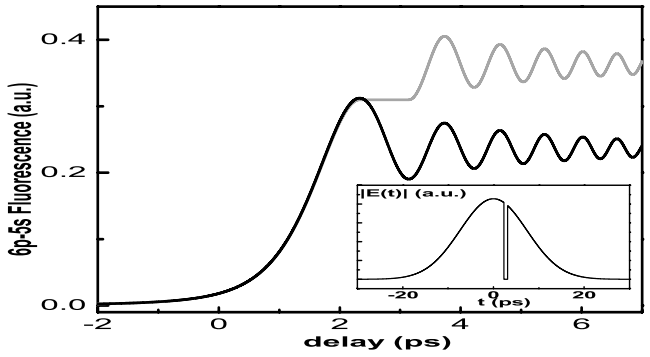

FIG. 3. Fresnel zone lens analogy: Excited transient population with unshaped chirped pump pulse (black line) and with calculated shaped pulse (solid gray line). Inset: Corresponding shaped temporal envelope of the pump pulse.

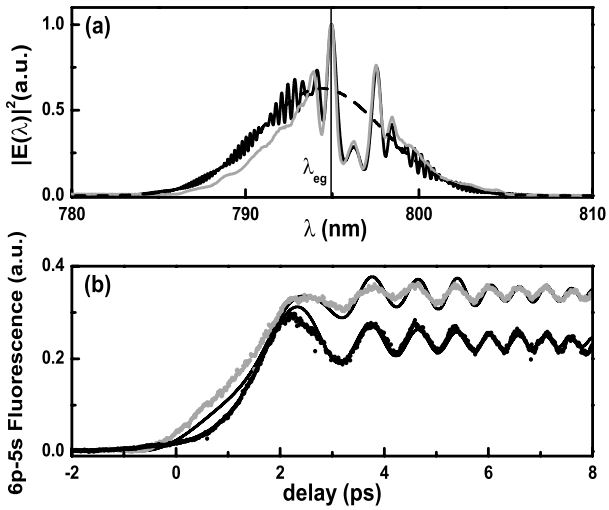

FIG. 4. (a) Calculated (black) and realized (gray) spectra. Original spectrum (dashed line). $\lambda_{eg} = 794.95$ nm. (b) Transient population measurements (dots) and numerical solution (line) obtained with the experimental spectrum (gray), together with the normal CT (black). Here $\phi'' = -4.9 \times 10^5$ fs^2 corresponding to $\tau_c \simeq 12.4$ ps.

this theoretical CT corresponds well to the experimental conditions. The amplitude normalization factor between experiment and simulation and the delay origin are kept identical from the fit of the normal CT. The theoretical shaped CT are, hence, obtained without any adjusted parameter. The theoretical curve reproduces perfectly all the experimental features. The contrast is very good, and the slope of the step is also very well reproduced. Clearly, the finite resolution of the pulse-shaping apparatus can be identified as the source of experimental imperfection.

Supplementary simulations explain that the disappearance of the "plateau" at the cutting time is due to the neglection of the residual, nonquadratic phase. Similar experiments have been performed to suppress the first increasing part in the oscillations leading again to a good agreement with theoretical simulations.

In conclusion, this experiment is the first demonstration of a pulse-shaping strategy where a complex shape (in-phase and amplitude) has been designed and applied to a simple system with an excellent agreement with predictions. We have manipulated the perturbative chirped excitation of a two-level system. In analogy to spatial Fresnel lenses, the temporal oscillation of the transient population was controlled both in phase and in amplitude. The results demonstrate that if the system is well known it is possible to predetermine the control mechanism and the exact corresponding shape and to apply it directly without feedback in open-loop coherent control.

We sincerely acknowledge F. Rosca-Pruna for her help at the final stage of the experiment and S. Zamith for fruitful discussions. We thank Professor K. L. Kompa for his generous support of this study. We also acknowledge financial support from the European Union (Contract No. HPRN-CT-1999-00129, COCOMO), the ESF ULTRA program, and Université Paul Sabatier APC program.

[1] H. Rabitz, R. de Vivie-Riedle, M. Motzkus, and K. Kompa, Science **288**, 824 (2000).
[2] L. Zhu, V. D. Kleiman, X. Li, S. P. Lu, K. Trentelman, and R. J. Gordon, Science **270**, 77 (1995).
[3] A. Assion *et al.*, Science **282**, 919 (1998).
[4] R. J. Levis, G. M. Menkir, and H. Rabitz, Science **292**, 709 (2001).
[5] C. J. Bardeen, V.V. Yakovlev, K. R. Wilson, S. D. Carpenter, P. M. Weber, and W. S. Warren, Chem. Phys. Lett. **280**, 151 (1997).
[6] D. Meshulach and Y. Silberberg, Nature (London) **396**, 239 (1998); N. Dudovich, B. Dayan, S. H. G. Faeder, and Y. Silberberg, Phys. Rev. Lett. **86**, 47 (2001).
[7] T. Hornung, R. Meier, D. Zeidler, K.-L. Kompa, D. Proch, and M. Motzkus, Appl. Phys. B **71**, 277 (2000).
[8] S. Vajda, A. Bartelt, E.-C. Kaposta, T. Leisner, C. Lupulescu, S. Minemoto, P. Rosendo-Francisco, and L. Wöste, Chem. Phys. **267**, 247 (2002).
[9] T. C. Weinacht, J. Ahn, and P. H. Bucksbaum, Nature (London) **397**, 233 (1999).
[10] A. P. Heberle, J. J. Baumberg, and K. Köhler, Phys. Rev. Lett. **75**, 2598 (1995).
[11] J. Kunde, B. Baumann, S. Arlt, F. Morier-Genoud, U. Siegner, and U. Keller, J. Opt. Soc. Am. B **18**, 872 (2001).
[12] J. L. Herek, W. Wohlleben, R. J. Cogdell, D. Zeidler, and M. Motzkus, Nature (London) **417**, 533 (2002).
[13] A. M. Weiner, Rev. Sci. Instrum. **71**, 1929 (2000).
[14] R. S. Judson and H. Rabitz, Phys. Rev. Lett. **68**, 1500 (1992); W. S. Warren, H. Rabitz, and M. Dahleh, Science **259**, 1581 (1993).
[15] T. Hornung, R. Meier, R. de Vivie-Riedle, and M. Motzkus, Chem. Phys. **267**, 261 (2001); H. U. Stauffer, J. B. Ballard, Z. Amitay, and S. R. Leone, J. Chem. Phys. **116**, 946 (2002); J. B. Ballard, H. U. Stauffer, Z. Amitay, and S. R. Leone, J. Chem. Phys. **116**, 1350 (2002).
[16] R. G. Brewer and R. L. Shoemaker, Phys. Rev. Lett. **27**, 631 (1971); B. Macke and P. Glorieux, Chem. Phys. Lett. **14**, 85 (1972); D. Grischkowsky, Opt. Commun. **18**, 69 (1976).
[17] S. Zamith, J. Degert, S. Stock, B. De Beauvoir, V. Blanchet, M. A. Bouchene, and B. Girard, Phys. Rev. Lett. **87**, 033001 (2001).
[18] R. Netz, T. Feurer, G. Roberts, and R. Sauerbrey, Phys. Rev. A **65**, 043406 (2002).
[19] N. Dudovich, D. Oron, and Y. Silberberg, Phys. Rev. Lett. **88**, 123004 (2002).
[20] B. Broers, L. D. Noordam, and H. B. van Linden van den Heuvell, Phys. Rev. A **46**, 2749 (1992).
[21] C. V. Bennett and B. H. Kolner, Opt. Lett. **24**, 783 (1999); L. Ménager, I. Lorgeré, and J. L. Le Gouët, Opt. Lett. **25**, 1316 (2000); L. Ménager, J.-L. Le Gouët, and I. Lorgeré, Opt. Lett. **26**, 1397 (2001).
[22] E. Riedle, M. Beutter, S. Lochbrunner, J. Piel, S. Schenkl, S. Spörlein, and W. Zinth, Appl. Phys. B **71**, 457 (2000).
[23] M. M. Wefers and K. A. Nelson, J. Opt. Soc. Am. B **12**, 1343 (1995).
[24] D. Zeidler, T. Hornung, D. Proch, and M. Motzkus, Appl. Phys. B S125 (2000).
[25] J.-P. Geindre, P. Audebert, S. Rebibo, and J.-C. Gauthier, Opt. Lett. **26**, 1612 (2001).