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## Coherent control approaches\*

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### Abstract

I review in this chapter the main approach to coherent control. The open-loop approach is emphasized. This approach is particularly suitable for simple systems (atoms here). It provides a fruitful basis which can be used to understand the effect of shaped pulses in more complex systems. Some examples using non-classical light as well as application for quantum information are also presented.

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## 1 Introduction

Use of optimally shaped pulses to guide and control the temporal evolution of a system has been an active field of research in the last decade after the initial proposal by Rabitz *et al* in 1992 [1, 2]. Recent experimental advances have led to successful control [3] of molecular ionization and dissociation, atomic and molecular fluorescence, clusters reactivity, the shape of Rydberg wave functions, excitations in semiconductors [4] but also of the energy flow in a biological system. These works were spurred by the technological breakthrough [5] for generating arbitrarily tailored pulses.

The effect of laser pulse shapes on the quantum system is related to the nature of the interaction. For a linear response of the system (one-photon transition in the weak field regime), the final outcome depends only on the spectral component at the resonance frequency and is therefore independent of the general pulse shape, and particularly of the spectral phases. This explains for instance why signals equivalent to wave-packet interferences could be observed with incoherent light as well as with ultrashort pulses. However, the temporal evolution towards the final state may depend strongly

on the pulse shape. A straightforward illustration of this statement is the non-resonant interaction which leads to transient excitation of the system, but to no final excitation.

For a non-linear interaction such as a multiphoton process or a one-photon transition in the saturation regime, the situation is drastically different. Many frequency components (if not all) contribute to the final state which is therefore strongly sensitive to shaping effects.

In the absence of predesigned control mechanisms only a closed loop scheme [1, 2] may be employed to find efficient pulse shapes [6]: 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 [7, 8, 9, 10]. This open-loop approach is well adapted to small systems for which theoretical predictions are reliable. It consists of reaching a specific goal (manipulation of the temporal response of a system excited by a light pulse) without any experimental feed-back. Physical analysis of the process allows to predetermine the theoretical pulse shape which leads to the desired result. It is then implemented experimentally.

This chapter presents a small overview of important results obtained in coherent control. Section 2 describes briefly the main features of shaped pulses and pulse shapers which are used throughout the chapter. The examples of the approach of open-loop control are given in section 3 for Coherent Transients observed during a one-photon excitation and in section 4 for final population control in two-photon absorption. Section 5 is dedicated to experiments using non-classical light while section 6 gives some references on quantum information schemes using shaped pulses.

## 2 Ultrashort shaped pulses

We describe in this section the basic tools used to modify the laser pulse shapes. The simplest way to shape ultrashort pulses consists in using passive dispersive devices. A pair of gratings used in double pass is an efficient way to generate pulses with strong chirps. The chirp of the pulse is varied by changing the distance between the gratings. Several experiments presented in this chapter have been performed with chirped pulses. Glass rods of strongly dispersive materials (SF10, SF58 ...) can also be used to generate strong fixed

chirps. The last decade has seen the development of active devices which offer wide pulse shaping capabilities. These pulse shaping techniques have been reviewed in details [5]. Due to their ultrashort duration femtosecond pulses are not easily shaped in the time domain. Thanks to the Fourier transform, the common way is to synthesize them in the spectral domain. The most usual device for both high fidelity and wide flexibility of shapes involves a pair of diffraction gratings and lenses arranged in a zero-dispersion 4-f line with a pulse shaping mask at the Fourier plane. This mask which can be a liquid crystal spatial light modulator (LC-SLM), acousto-optic modulator or deformable mirror, modulates the spectral phase and/or amplitude. Shaping without Fourier transform optics is also possible by using an actively controlled acousto-optic programmable filter [11]. In the following experiments, the pulse shaper devices are principally based on a 4f line using cylindrical mirrors instead of lenses to limit chromatic aberrations. Liquid crystal modulators are placed in the Fourier plane. Depending on the experiment, 128 pixels phase-only or phase and amplitude or 640 pixels amplitude and phase are used [12]. The effect of the pulse shaper is to multiply the input electric field  $E_{in}(\omega)$  by a filter function  $A(\omega) e^{i\phi(\omega)}$  :

$$E_{out}(\omega) = E_{in}(\omega) \times A(\omega) e^{i\phi(\omega)} \quad (1)$$

The complete characterisation of ultrashort pulses is an active field of research. One possibility is to use a method "with reference" such as cross-correlation or spectral interferometry. If no reference is available then several self-referenced methods have been proposed. These are based on two main approaches. The FROG (FRequency Optical Gating) uses a gate shorter than the structures of the pulse. The pulse shape is deduced from iterative analysis. It leads to many derivatives.[13]. The SPIDER (Spectral Phase Interferometry for Direct Electric-field Reconstruction) combines spectral interferometry with frequency shearing. In this case contrary to the previous one no algorithm is required. The phase of the electric field is directly obtained after only Fourier transforms and filtering [14]. Recently an alternative method has been proposed based on the coherence of the atoms in a light-matter interaction [15].

### 3 One-photon transition

When the interaction between the electromagnetic field and the matter is in the linear regime, the final state populations can be entirely deduced from the power spectrum. This is for instance the case for a one-photon transition in the weak field regime. However, the phase of the wave function is sensitive to the various phases of the electric field. This can have important consequences for applications where a subsequent excitation is performed, in particular when coherent superpositions are involved. The transient evolution of excited state population is also strongly dependent on the detailed laser shape. As an intuitive illustration of this statement, the transient response to a non-resonant excitation follows the electric field pulse envelope, independently of its spectrum. For instance, simply changing the pulse duration will change this transient response.

A resonant interaction leads to radically different behaviors. FT limited pulses produce a step-wise excitation in the weak-field, and Rabi oscillations in the intermediate and strong field regime. Chirped pulses produce a total population inversion in the strong field with a final state robust with respect to small variations of laser parameters [16]. Chirped pulses in the weak field lead to a less intuitive behavior which illustrates 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 [8]. These interferences are named coherent transients and have been experimentally observed and manipulated([10]).

The high sensitivity of Coherent Transients to slight modifications of the laser pulse opens possibilities to use CT measurements as a characterization of the laser pulse[17]. One difficulty resides in the fact that only the part of the pulse after resonance leads to oscillations which can be used to determine the shape. Another difficulty is that the measured quantity is related to the probability whereas the the probability amplitude which is proportional to the integral of the laser electric field is not directly measured [15].

These difficulties can be overcome by combining several measurements. Several CT measurements are combined, each with a sequence of two pulses. This sequence consists of a reference pulse which creates an initial population in the excited state, and an "unknown" pulse whose effect on the quantum state is to be measured in real time. The first pulse creates an initial population in the excited state so that the corresponding dipole beats with the

whole of the second (unknown) pulse. A second measurement is performed after adding a  $\pi/2$  phase shift to the second pulse. This provides in-phase and in-quadrature measurements from which the real-time evolution (*during the laser interaction*) of the quantum state can be deduced.

In common with quantum state holography [18], the quantum state created by the unknown pulse is determined by direct interferometric comparison with a reference quantum state. However, in our method, the unknown quantum state is measured at any time during and after its creation[19].

## 4 Two-photon transition with classical light

Two-photon transitions represent the lowest-order non linear interaction and they are thus chosen as an illustration of some control schemes. In particular quantum interferences between the various paths leading to the same final state are allowed [20]. These schemes have been well studied in atoms to understand several possibilities of controlling these interferences ( See for example[21]). In the case of two-photon transitions two cases are easily distinguishable depending on whether intermediate states are close to resonance [22] or not [7, 23]. Let us note that in the case of non-resonant intermediate states, two-photon transitions and a fortiori multiphoton transitions can be reached by many routes through a continuum of virtual levels. In the case of a short pulse excitation, due to the corresponding broad bandwidth it is instructive to examine first the summation over the intermediate levels. At that point, one has to distinguish the case where these are close or far from resonance.

### 4.1 Weak-field two-photon transition with no resonant intermediate state

In contrast to a single photon transition, all frequency components of the pulse contribute to the transition. More precisely each pair of photons of frequencies  $(\omega_i, \omega_j)$  contained in the pulse spectrum which verify  $\omega_{eg} = \omega_i + \omega_j$  contribute to the TPA. The two-photon transition probability can therefore be controlled by tailoring the spectral phase of a single pulse. It is easy to demonstrate that the flat phase ( $\phi(\omega) = 0$ ) maximizes the TPA. One well-known example concerns the second harmonic generation in a non linear crystal : the pulse which maximizes such signal is the transform limited

pulse [24]. Since pulse shaping cannot help improve the TPA efficiency when no intermediate state is involved, it can be used to reduce it by introducing destructive interferences between the various paths involved. This approach, besides its fundamental interest, can in practice be a better alternative than simply reducing the laser intensity, since it can allow to control selectively a particular transition.

Silberberg *et al* have demonstrated such a control by tailoring the pulse spectral phase [7]. A phase-only 128 pixels SLM pulse shaper was used. This does not affect the power spectrum. A spectral  $\pi$ -step applied at resonance provides a simple antisymmetric shape for which the TPA rate is unchanged. Shifting the position of this  $\pi$ -step changes progressively the balance between negative and positive contributions of the electric field[23]. For a particular position, they counterbalance exactly each other and the TPA rate vanishes. These general principles hold for multiphoton absorption including other multiphoton processes[3] with application to gas-phase molecules or solid state systems, to multiphoton microscopy and Coherent Anti-Stokes Raman scattering.

## 4.2 Weak-field two-photon transition with a resonant intermediate state

The efficiency of a multiphoton process is greatly enhanced when the intermediate state is nearly resonant. This is in fact easier to achieve with ultrashort pulses, thanks to their broad spectrum [25]. This can be the case with several atomic or molecular excited electronic states. This is indeed intrinsically the case of vibrational levels in a molecule since they are nearly equidistant. For instance, exciting molecules towards a specific highly excited vibrational state can be efficiently achieved with a multiphoton transition using photons tuned close to the vibrational frequency [26] whereas direct single-photon excitation is almost forbidden by selection rules. Therefore stepwise excitation up the vibrational ladder is required. Several experiments have been performed using far-infrared lasers ([27]). However due to the complexity of molecular systems, combined with the experimental difficulties inherent to the excitation and characterization of infrared transitions, it is instructive to study extensively a theoretically and experimentally simpler system, an anharmonic quantum ladder (with two steps) in an alkali atom (Rb or Na) [28, 29]. In the presence of a resonant intermediate states  $|i\rangle$  the sequential



paths opened by the presence of the intermediate levels have a crucial role. In this case it is possible to enhance the transition probability by shaping the excitation pulse. In a first spectacular example, a highly counterintuitive result is obtained by Silberberg's group : Blocking parts of the spectrum of the exciting pulse (corresponding to destructive interferences) which reduces the power by 71% leads to an enhancement of the TPA rate by a factor of 2. In a second example, applying a phase shift of  $\pi/2$  to a spectral window inverts the sign of  $\tilde{E}(\omega)\tilde{E}(\omega_{eg}-\omega)$  in order to compensate for the sign change at resonance. This maximizes the enhancement of the TPA rate by a factor of 7[22].

Another simple pulse shape is obtained by simply chirping the pulse with a quadratic spectral phase [29, 25]. This provides a symmetric phase function which is expected to induce significant control capabilities. This approach offers several advantages due to the simplicity of the theoretical treatment and of the experimental set-up. Very nice quantum interferences have been obtained with a contrast around 80%.

## 5 Coherent control with non-classical light

Light generated by a down conversion process can be broadband just like a short pulse, yet it is not coherent. Still, the inherent correlations between the signal and idler waves offer new possibilities, as recently demonstrated by Silberberg [30]. When broad-band light is generated via down conversion of a narrow-band pump, it combines two seemingly incompatible properties: It performs two-photon absorption with the efficiency and temporal characteristics of a femtosecond pulse, but it is sensitive to the tuning of the wavelength of the pump as if it was a cw experiment [30]. Coherent control schemes have been also implemented with this high power, broadband squeezed vacuum [31].

Entangled photons often behave as a single particle. If they arrive as single pairs, they perform nonlinear interactions at a rate that is linear with their flux. Silberberg *et al* have generated a very high flux of entangled photons by very broad down conversion. Even though they had some 1012 photon pairs per second, they still had less than one pair per mode. An extremely linear dependence of a nonlinear interaction - sum frequency generation - on the number of pairs has been thus demonstrated [32]. The SFG could also be controlled and shaped using frequency domain manipulations

of the relative phases of the photons, just like shaping femtosecond pulses.

These results open a new field of applications combining coherent control with quantum optics.

## 6 Coherent control approach for quantum information

Finally, several examples using the coherent control approach in the field of quantum information, are briefly presented. The first one demonstrates the implementation of Grover's algorithm using Rydberg states[33].The second one presents the implementation of an AND gate using rovibrational states [34]. In both cases, quantum states are used as a register, shaped pulses are used as "write" pulse while another probe pulse is use to read the data.

In our group another approach has been proposed : The predissociation of a diatomic molecule after excitation by an ultrashort laser pulse leads to a succession of partial nuclear wave packets on the dissociative potential curve. Matter wave interferences result from their evolution. We have shown theoretically how control pulses inducing Rabi oscillations on the electronic atomic part of the wave function can change totally the shape of the nuclear interference pattern. This is a consequence of the entanglement between the electronic and nuclear parts of the wave function. This leads to application in information storage [35].

Finally a new scheme based on Gauss sum to factor numbers has been implemented using shaped pulses. Indeed, the frequency component of the electric field corresponding to a sequence of appropriately shaped femtosecond pulses is determined by a Gauss sum which allows us to find the factors of a number [36].

## 7 Conclusion

Coherent control has been an extremely active field of research in the last decade. Technological implementations in terms of shaped pulses, tunability and easy-to-use pulses have allowed all these progresses. Very recent results combining coherent control approach and quantum optics seem to be really promising and could lead to real progress in term of understanding of quantum properties of light as well as quantum information.

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