

Characterisation of Ultrashort pulses



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To my Mum.

ABSTRACT

The main topic of this thesis is the development of new characterization techniques for Ultrashort pulses with particular focus on pulses with complex spectra, often obtained with light pulse synthesisers. More in detail, I have addressed the longstanding problem of retrieving the spectral phase of a pulse which exhibits a spectral gap, i.e. a spectral null larger than any of the spectral features of the pulse. The major challenge in this scenario is attaining a reliable reconstruction of the relative spectral phase between the spectral components. I discuss the different approaches that I have employed in order to achieve this objective.

The first result has been the realisation of a low-power XFROG for the characterisation of a coherent pulse synthesis source developed by Prof. D. Reid's group at Heriot-Watt University. We used it to characterise, compress, and synchronise the individual components of the source, however the relative phase remained elusive. In order to solve this problem, a radically new characterisation technique needed to be implemented. This consisted in the development of a new algorithm (MICE), which enabled the mutual characterisation of multiple fields, and in a new experimental implementation of the well-known SPIDER technique (SPICE). Both the algorithm and the new technique have been tested experimentally in a proof-of-principle experiment which constitutes the main result of this work. The appeal of these solutions for mastering light synthesisers will likely stimulate further refinements towards making it the standard technique for complex spectra.

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INTRODUCTION

Attaining laser pulses with the shortest possible duration has been an ongoing quest in the scientific community for the last few decades, which saw its first results with the discovery of mode-locked lasers [1–3]. The interests of this endeavour is both of a fundamental character and of a practical one, as ultrashort lasers enable applications in studying and monitoring of those phenomena too fast to be directly probed via electronic scopes, such as the motion of molecules in biological environments [4], chemical reactions [5] and processes in plasma [6]. In this respect, time-resolved pump-probe experiments performed by ultrashort pulses are probably the most diffused technique for inferring information at such short timescales.

Fig. 1.1 gives us a neat explanation of how a pump-probe experiment works. If we want to study the dynamics of a tennis shot, first, we need to send the ball to trigger a response from the tennis player: this is exactly why a pump is needed. We can then probe the shot movement by taking pictures at regular intervals and, by analyzing them, information can be gathered on the ball's trajectory and the underlying forces. In order to successfully extract such information the choice of the sampling frequency, and consequently the exposure time of each picture, is crucial: we need to avoid undersampling as well as keeping the camera on for too long. The former would lead



Fig. 1.1 An example of a time-resolved image of Roger Federer at 2012 Olympics Games. Courtesy of Tullia Cecchetti.

to loss of information, while the latter would result in blurry pictures. When dealing with faster dynamical processes than Roger Federer shooting, the basic principles remain identical, however more advanced and faster pump-probing techniques need to be implemented to meet the new, more demanding requirements. Outside our analogy, in a time-resolved pump-probe experiment, a dynamical system is driven by a pump in the form of a laser pulse, and then probed by a second pulse at regular time intervals. Timing and duration of such probes are dictated by the specific process. Since molecular motion occurs over a fraction of a picosecond, the considerations above lead naturally to the adoption of few femtoseconds probes. Nowadays few-optical-cycle pulses are routinely produced directly from laser oscillators [7, 8], by spectral broadening within a Hollow-Core Fibre (HCF) [9, 10], by filamentation in gases [11] and by optical parametric amplifiers (OPA) [12]. The level of control we can achieve via these pulses is however conditioned by the level of control we have on the pulses themselves: their time duration needs to be carefully assessed if we intend to make use of them as probes of physical phenomena. Ultrafast metrology techniques have been developed and they are nowadays commonplace diagnostics; they provide an indirect measurement for the duration of the pulses by recovering

their spectral phase, which, together with the easily-measurable spectral amplitude, supplies the information needed to reconstruct the pulses' temporal profile. Over the years different techniques have been introduced [13–15] to respond to the demands presented by the new sources; for example, tailored techniques have been conceived for weak [16, 17], ultrabroadband [18–20], and structured pulses [21–23].

The quest for the shortest pulses has now progressed well beyond the femtosecond regime via the introduction of High Harmonic Generation (HHG) sources [24–26]. Here an intense laser pulse is focused onto a gas jet, producing a strongly nonlinear interaction in the ionization regime; this leads to the generation of harmonics of high order, reaching the extreme ultraviolet (XUV) spectral region. Higher frequencies are necessary to act as carrier for shorter pulses since a full optical cycle in the visible takes a few femtoseconds. A complementary approach consists in coherently combining femtosecond lights sources to produce sub-single-cycle waveforms [27] tailored to different uses (as for example, acting as seed to generate isolated attosecond pulses with HHG [28–30]). These sources require a level of control beyond the mastering of each individual source; in particular, the spectrum of these light synthesizers often presents well-separated spectral components, as the consequence of reaching for large bandwidths and tailoring the temporal waveform at the same time. Virtually all techniques for ultrafast metrology developed so far rely heavily on a compact spectrum¹. Adapting them to overcome these limitations, demands a careful rethinking of the experimental design and the methods of analysis applied in conventional techniques. The reward would be a superior level of control of the waveform engineering and the opening of novel capabilities for dynamical probing and control. This thesis aims at developing a new metrology technique capable of measuring pulses which exhibit such *spectral gaps*, that is, spectral regions with zero intensity between spectral components. The first attempt I pursued has been to

¹a spectrum that does not have multiple separated spectral components.

adapt an existing technique, namely XFROG, within a collaboration with the group of Prof. Reid at Heriot-Watt University. Prof. Reid's group had developed a coherent synthesis source with spectral gaps, that needed to be optimized. Despite the experiment proved to be limited by the available laser power, our characterization setup was able to deliver key information for the simultaneous compression of the individual spectral components. The research for a new technique went alongside with the development of a new algorithm on which I have worked together with Dr. C. Bourassin-Bouchet and Dr. M. M. Mang, called Mutual Interferometric Characterization of Electric-fields (MICE). This became an indispensable tool for the success of the new technique itself. The algorithm allows for the simultaneous retrieval of the spectral phase of more than one pulse at the same time. Finally the new technique, which was named SPectral-gap Immune Characterization of Electric-fields (SPICE), was developed and tested in a proof-of-principle demonstration. It provides a way to retrieve the phase of a pulse with spectral gaps, as long as a pulse with a continuous spectrum covering those gaps (whose phase can be unknown) is available. I have performed a proof-of-principle experiment for the SPICE technique with one tunable gap in which the relative phase of two separated components was correctly retrieved. The experiment presented some complications which will be thoroughly discussed, but nevertheless provided a validation for the new SPICE technique. While in this work SPICE has been applied to temporal characterization of visible pulses, its capabilities go beyond that and can further be developed to lead, together with MICE, to a full all-optical characterization of XUV pulses.

1.0.1. THESIS OUTLINE

In this thesis I present the results obtained in the process of developing the SPICE technique.

Chapter 2 reviews of all the concepts and terminology commonly used in nonlinear optics that will be used in this thesis.

Chapter 3 gives an overview of the existing metrology techniques. The description and analysis of the MICE algorithm is also included.

Chapter 4 discusses the results obtained within the Heriot-Watt collaboration using the XFROG setup to simultaneously retrieve the phases of the parent pulses of the coherent pulse synthesis source developed in Prof. Reid's group.

Chapter 5 provides the theoretical framework for the SPICE technique and the simulations carried out to test it against noise and space-time coupling.

Chapter 6 presents the results obtained for the proof-of-principle experiment of SPICE.

Chapter 7 outlines the main conclusions for the work here presented and provides insights on the future developments.

CHAPTER 2

BACKGROUND

Have you ever wondered, well I have.
About how when I say, say red, for example.
There's no way of knowing if red
Means the same thing in your head
As red means in my head.
When someone says red.

Matilda, Tim Minchin

In this chapter I introduce the fundamental concepts that will be necessary for the description of the experiments performed and algorithms developed in this thesis. After defining what an ultrashort pulse is, and all its relevant characteristics I give a brief review of new ultrashort pulses sources in which the techniques developed in this work currently find their main application.

2.1. DESCRIPTION OF ULTRASHORT PULSES

Light pulses are described by the *analytic electric field*, which can be written as a real envelope function $A_t(t)$ modulated by a fast carrier wave:

$$E(t) = A_t(t)e^{(i\phi_t(t)-i\omega_0 t)}, \quad (2.1)$$

where ω_0 is the carrier frequency and $\phi_t(t)$ is the time-dependent phase. If we consider the electric field as a complex signal, it is immediate to consider that, in order to uniquely determine it, an amplitude and a phase need to be specified. As it will be detailed in the next chapter this is experimentally achievable if we consider the electric field in its frequency representation, which is obtained by fourier transforming (2.1):

$$\tilde{E}(\omega) = \int_{-T}^T E(t)e^{i\omega t} dt = A_\omega(\omega)e^{i\phi_\omega(\omega)}, \quad (2.2)$$

where $[-T, T]$ is the compact support of $E(t)$, $A_\omega(\omega)$ is the spectral amplitude, and $\phi_\omega(\omega)$ is the spectral phase. Here we adopt a description of the spectrum in terms of positive frequencies only.

The amplitude is linked to the spectrum of the pulse via $S(\omega) = \langle |A_\omega|^2 \rangle$ where the brackets indicate the average over time; the spectral phase can be written as a Taylor expansion around ω_0 :

$$\begin{aligned} \phi_\omega(\omega) = \sum_{n=0}^{\infty} \frac{1}{n!} \phi_\omega^{(n)}(\omega_0) (\omega - \omega_0)^n = \phi_\omega(\omega_0) + (\omega - \omega_0) \left(\frac{\partial \phi_\omega}{\partial \omega} \right) \Big|_{\omega_0} + \\ + \frac{1}{2} (\omega - \omega_0)^2 \left(\frac{\partial^2 \phi_\omega}{\partial \omega^2} \right) \Big|_{\omega_0} + \quad (2.3) \\ + \frac{1}{6} (\omega - \omega_0)^3 \left(\frac{\partial^3 \phi_\omega}{\partial \omega^3} \right) \Big|_{\omega_0} + \dots \end{aligned}$$

where $\phi_\omega^{(n)}$ is the n -th derivative of ϕ_ω . Equivalently the temporal phase can also be expressed as a Taylor expansion:

$$\phi_t(t) = \sum_{n=0}^{\infty} \frac{1}{n!} \phi_t^{(n)}(t_0) (t - t_0)^n, \quad (2.4)$$

where t_0 is a reference time. Each spectral phase derivative and its temporal counterpart has a physical meaning which is summarized in Fig. 2.1.

The zeroth order term in the spectral domain $\phi^{(0)}(\omega_0)$ is an absolute spectral phase. It is equivalent to the zeroth order term in the temporal domain which represents the *carrier-envelope phase* (CEP), which consists in the phase difference between the envelope and the carrier wave, as pictured in fig 2.1 b) where two identical pulses are shown with two different CEP phases. The zeroth order phase is irrelevant when dealing with many-cycle pulses but becomes increasingly important with few-cycle and synthesized sub-cycle pulses [31] or when dealing with phenomena which are sensitive to the oscillating field, as for example High Harmonic Generation (HHG) [24]. The linear term in the spectral domain $\phi^{(1)}(\omega_0)$, is the group delay (GD), which corresponds to the delay in time of the central wavelength. The group delay can be

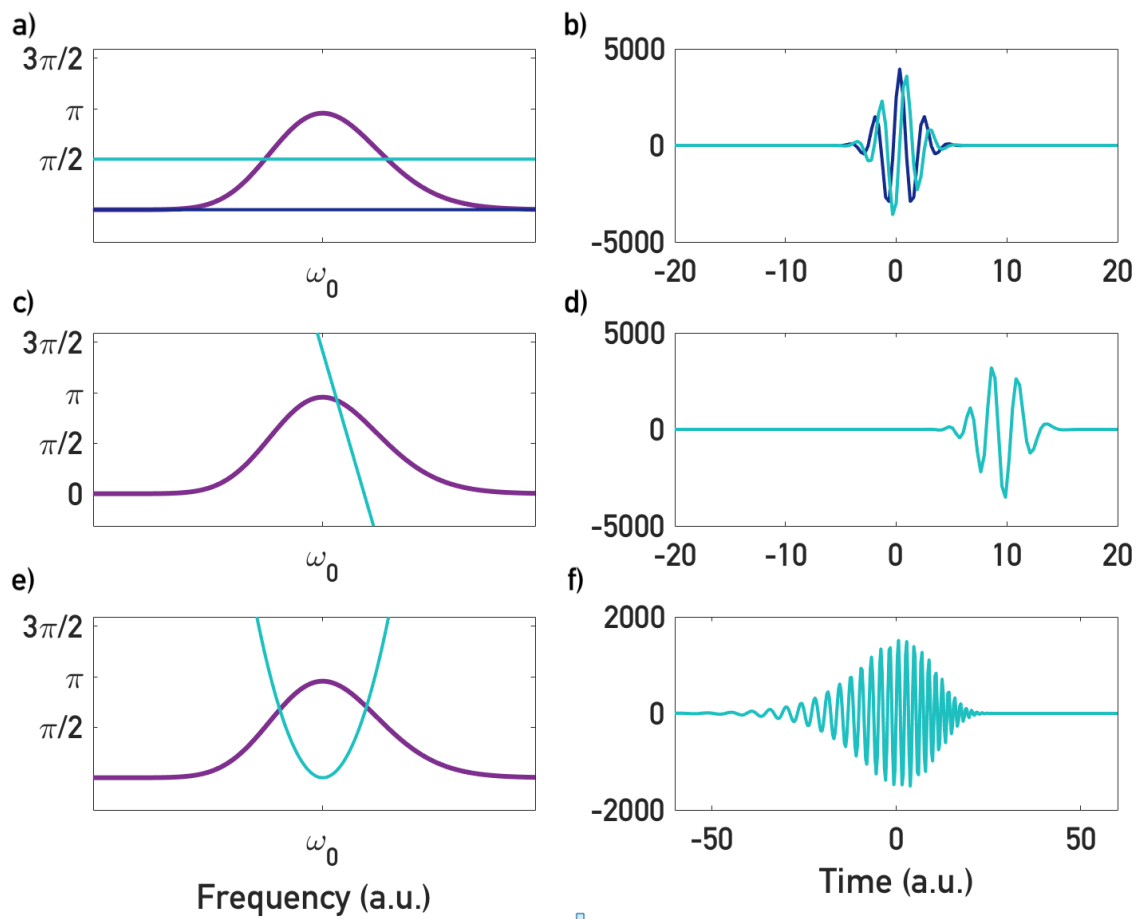


Fig. 2.1 Phase terms. The column on the left shows the Spectrum and spectral phase; the column on the right shows the respective electric field in time. The spectrum is the same in all the instances, while the phase changes from 0th order (first row) to 1st order (middle row) to 2nd order (bottom row).

dependent on the frequency. In this case also a second order phase derivative is present, the GDD, group delay dispersion $\phi^{(2)}(\omega_0)$. If the group delay is frequency dependent, it means that different frequencies will arrive at different times, resulting in a temporal broadening of the pulse, in which case the pulse is said to be chirped. Higher-order terms can lead to nonlinear chirps and are generally referred as TOD (third order dispersion) and FOD (fourth order dispersion) .

In the case where the GDD and all higher orders are zero, the pulse is said to be bandwidth limited or Fourier transform limited and it has the shortest duration possible for a given optical spectrum. If that is the case, for a pulse with a spectral full width at half maximum (FWHM) given by $\Delta\omega$ and a temporal duration given by the temporal FWHM Δt , the time bandwidth product holds:

$$\Delta\omega \cdot \Delta t = \alpha, \quad (2.5)$$

where α is order 1, and its actual value is determined by the envelope shape. As an example, for a Gaussian envelope the value of α is $\alpha 0.441$.

2.1.1. SPACE-TIME COUPLING

Up to now no information has been given regarding the spatial part of the pulse as the time and spatial components of a light pulses are usually independent and thus in most cases for a full description of the temporal profile there is no need to infer any condition on the spatial part and the complete field can be factorized into:

$$E(x, y, t) = E_s(x, y) \cdot E_t(t). \quad (2.6)$$

where x and y are the directions orthogonal to the propagation direction. However there are instances in which this is not the case, as during propagation correlations

can arise between the spatial and temporal profile of a pulse, that is, we are in the presence of Space-Time coupling (STC) [32].

One of the most common examples of STC is given by pulse front tilt (PFT). PFT consist in a rotation of the pulse group front¹ compared to the plane orthogonal to the propagation direction. It has been shown that such a behaviour can arise either due to angular dispersion [33, 34] or to the presence of temporal and spatial chirp at the same time [35].

In [35] Akturk et al. show how the PFT can be characterized by the derivative of the pulse front arrival time t_0 with respect to the position x :

$$p = \frac{dt_0}{dx}. \quad (2.7)$$

In the general case the parameter p is composed of two terms:

$$p = p_{AD} + p_{SC+TC}, \quad (2.8)$$

where the first term is due to the angular dispersion and is given by $p_{AD} = k_0\beta$, where k_0 is the wave number and $\beta = \frac{d\theta_0}{d\omega}$ is the angular dispersion; the second term is given by the combination of the spatial and temporal chirp and is equal to $p_{SC+TC} = \phi^{(2)}\nu$, where $\phi^{(2)}$ is the GDD and $\nu = \frac{d\omega_0}{dx}$ is the spatial chirp rate.

The expression of a pulse with PFT in the presence of spatial chirp will then be given by:

$$E(x, y, \omega) = E_{xy}(x, y)E_{\omega}(\omega - \nu x)e^{-ipx\omega}. \quad (2.9)$$

¹pulse group front is defined as the contour of equal intensity for a fixed time t .

In this thesis the pulses have been always kept STC-free apart from particular instances in which experimental constraints did not allow for alternatives. This will be detailed thoroughly in Chapter 5 and 6.

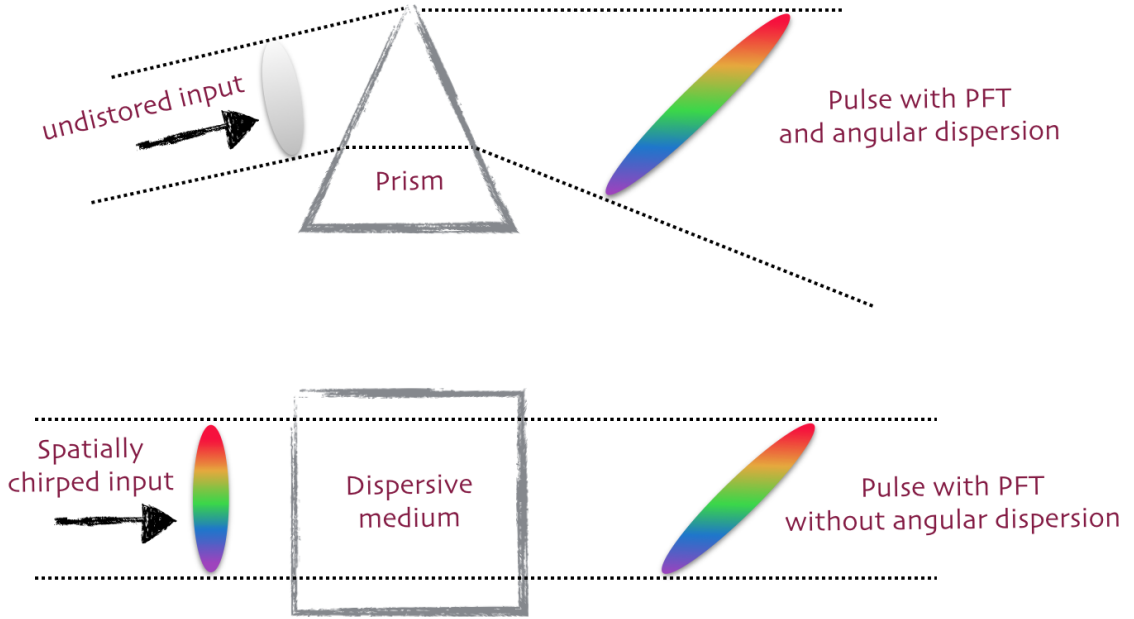


Fig. 2.2 Pulse front tilt generation. Upper image: the beam goes through a prism and the pulse group front is rotated due to angular dispersion. Lower image: a previously spatially chirped beam goes through a dispersive medium, which causes the frequencies to shift in time due to GDD, resulting in PFT.

2.2. PROPAGATION THROUGH A MEDIUM

When propagating in a dielectric medium, the spectral phase will be affected by dispersion, i.e. the wavelength-dependent refractive index $n(\lambda)$ [36], which for many material can be described by the Sellmeier equations [37]:

$$n(\lambda) = \sqrt{1 + \sum_j \frac{B_j \lambda^2}{\lambda^2 - C_j}}, \quad (2.10)$$

where the coefficients representing the coupling strength (B_j) at a wavelength $\sqrt{C_j}$, are fitted empirically.

This will result in the pulse being affected by chromatic dispersion. Each term of the spectral phase can be written in terms of the refractive index if one considers that the accumulated phase after the propagation in a medium can be written as a product of the frequency-dependent wave vector $k(\omega)$ and the length of the medium L [38]

$$\phi(\omega) = k(\omega) \cdot L = \frac{n(\omega)\omega}{c}L. \quad (2.11)$$

If the refraction index is varying slowly enough, a Taylor expansion can be made, obtaining:

$$k(\omega) \simeq k(\omega_0) + (\omega - \omega_0) \left(\frac{\partial k}{\partial \omega} \right) \Big|_{\omega_0} + \frac{1}{2}(\omega - \omega_0)^2 \left(\frac{\partial^2 k}{\partial \omega^2} \right) \Big|_{\omega_0}. \quad (2.12)$$

The zeroth order term $k(\omega_0)$ is linked to the *phase velocity* v_ϕ via:

$$v_\phi = \frac{\omega_0}{k(\omega_0)} = \frac{c}{n(\omega_0)}, \quad (2.13)$$

while the first order is linked to the *group velocity* v_g via:

$$v_g = \frac{1}{k^{(1)}} = \left(\frac{d\omega}{dk} \right) \Big|_{\omega_0}. \quad (2.14)$$

The second order term is given by:

$$k^{(2)} = \frac{d^2 k}{d\omega^2} = \frac{d}{d\omega} \left(\frac{1}{v_g(\omega)} \right), \quad (2.15)$$

corresponding to the case where the group velocity is frequency-dependent, that is the *group velocity dispersion* (GVD). By multiplying these terms by the length of

the medium L , the first order becomes the time delay and the second order becomes the GDD we have seen in Section 2.1.

It is thus commonplace that by propagating through a medium, an ultrashort pulse will undergo a consistent temporal broadening due to the acquired spectral phase which has then to be taken into account. There are in fact different systems which can counteract the propagation effect by providing opposite dispersion to compress the pulse back to its Fourier limited value. Among the many compression techniques we consider dispersion compensation via a prism pair [38–40], which will be used later in Chapter 4. This technique is based on geometric dispersion, accomplished by utilizing wavelength-dependent paths: when a couple of prisms is set as in Fig. 2.3, the angular dispersion due to the first prism translates into different path-lengths for different wavelengths in the second prism: the red components will be less deflected by the first prism and this will result in a longer path inside the second prism compared to the blue components, inferring *negative dispersion* on the pulse. Passing only once through the prism pair however results in pulse front tilt and spatial dispersion so it is preferable to adopt a double pass geometry as of Fig. 2.3. It must be noted that this type of compressor adds also third-order dispersion which cannot be decoupled from the second order and is dependent on the length between the prisms and of the specific prism material.

Other compressor techniques include a pair of parallel gratings [41], chirped mirrors [42, 43], reflection grisms [44], or shapers like 4-f lines with a spatial light modulator (SLM) in the Fourier plane [45–47] which can be combined and tailored for specific compression needs.

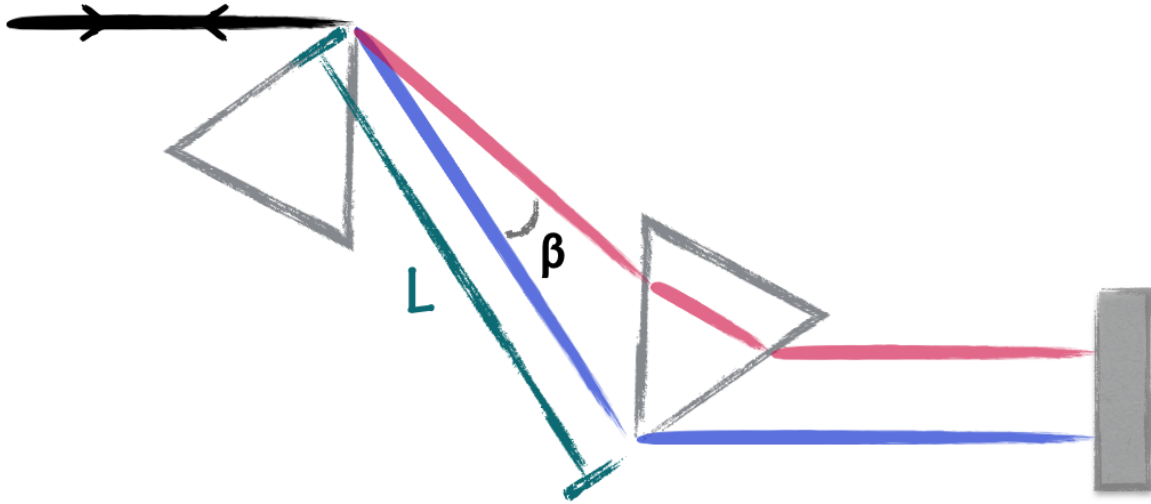


Fig. 2.3 Prism compressor. Scheme of a double pass prism compressor with beam deviation angle β and apex separation L .

2.2.1. NONLINEAR PROPAGATION

The response of the medium to the electric field of the propagating pulse is given by its polarisation P . The polarisation is the density of the induced electric dipole moments of the atoms inside the material due to the applied oscillating electric field. The nature of the interaction will depend linearly on the E -field strength: for weak fields, the oscillations are small and will result in a linear dependence of the polarisation with electric field strength; when the field strength increases the linear assumption does not hold and higher order terms need to be taken into account. This is also due to the fact that for higher energies the electrons potential cannot be considered harmonic anymore. The polarisation can thus be written as a power series expansion:

$$\vec{P} = \epsilon_0 \left(\chi^{(1)} \vec{E} + \chi^{(2)} \vec{E}^2 + \chi^{(3)} \vec{E}^3 + \dots \right) \quad (2.16)$$

where $\chi^{(n)}$ is the n-th order susceptibility and ϵ_0 is the free-space permittivity.

The wave equation given by:

$$\left(\nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) \vec{E} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \vec{P}}{\partial t^2} \quad (2.17)$$

will then become nonlinear. If the electric field strength is relatively small compared to the binding field of the atom² the propagation will be affected by different order nonlinear phenomena depending on the type of material considered. If the intensity is higher, strong fields physics phenomena such as HHG [24] will then occur due to the ionization of the bound electrons.

Here I briefly report the nonlinear effects which will be mentioned later in the thesis: sum frequency generation (SFG) and self-phase modulation. For an accurate description of nonlinear phenomena see [48].

2.2.2. SUM FREQUENCY GENERATION

When propagating through a noncentrosymmetric crystal³ as for example a BBO (β barium borate) crystal, the second order susceptibility $\chi^{(2)}$ can be non zero and second order nonlinear phenomena can occur.

If two input fields with frequencies ω_1 and ω_2 overlap on a nonlinear crystal two photons can annihilate to produce a third photon at ω_3 . In the process both energy and momentum are conserved, thus the relations $\omega_3 = \omega_1 + \omega_2$ and $\vec{k}_3 = \vec{k}_1 + \vec{k}_2$ need to hold. A graphic representation of the SFG process is given in Fig. 2.4. If the two input fields have the same frequency ω_1 then the resulting field will be at $\omega_3 = 2\omega_1$ and the process takes the name of second harmonic generation (SHG) .

²For linearly polarised light the field intensity needs to be $I \ll 10^{15} \text{ W cm}^{-2}$.

³A crystal is said to be noncentrosymmetric when it does not display inversion symmetry.

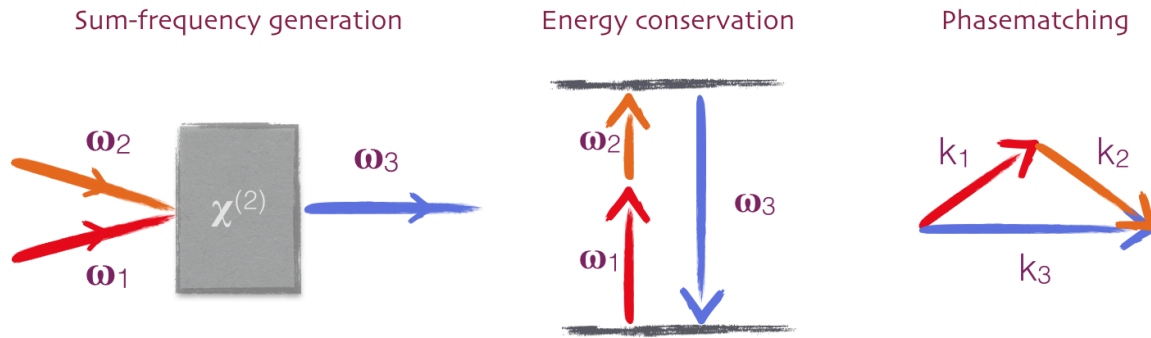


Fig. 2.4 Sum-frequency generation. Two beams with frequencies ω_1 and ω_2 combine on a nonlinear second order crystal resulting in a signal at $\omega_3 = \omega_1 + \omega_2$ (right). The process must abide both energy conservation (centre) and momentum conservation (left).

The momentum conservation condition is commonly called *phase matching*. In terms of refracting index it can be rewritten as:

$$\frac{n(\omega_3)\omega_3}{c} = \frac{n(\omega_1)\omega_1}{c} + \frac{n(\omega_2)\omega_2}{c}. \quad (2.18)$$

This can be hard to be achieved in materials with normal dispersion, where the refractive index increases with the frequency: if we consider the simple case in which $\omega_2 = \omega_1$, then from (2.18) $n(\omega_1) = n(2\omega_1)$ which can not happen if the refractive index increases monotonically with ω . In order to achieve the phase-matching condition of (2.18), the common technique is to use birefringent materials in which the refractive index depends also on the polarisation of the optical radiation. In uniaxial crystals there is one particular direction (extraordinary axis) in which the refractive index assumes the values $n_e(\omega)$, while on the other two directions (ordinary axes) the refractive index will be $n_o(\omega)$. Uniaxial crystals can be either positive (if $n_e > n_o$) or negative (if $n_o > n_e$), which will result in different phase-matching configurations. If

Type	Positive ($n_e > n_o$)	Negative ($n_o > n_e$)
Type I	e e $\rightarrow$ o	o o $\rightarrow$ e
Type II	o e $\rightarrow$ o	o e $\rightarrow$ e

Table 2.1 Phase matching scheme for uniaxial crystals

the propagation happens on a generic direction, forming an angle θ with respect to with the extraordinary axis, it will be subject to a refractive index given by:

$$\frac{1}{n(\theta, \omega)^2} = \frac{\sin^2 \theta}{n_e(\omega)} + \frac{\cos^2 \theta}{n_o(\omega)}. \quad (2.19)$$

This relation makes it possible, by tuning the angle θ , to achieve phase matching for the desired frequencies. Depending on the polarisation of the beams different phase-matching conditions can be reached: if the two input beam have the same polarisation and the outcoming beam has the orthogonal one, the phase matching is said to be of **Type I**; if the input beams have orthogonal polarisations then the phase matching is said to be of **type II**. The possible combinations for positive and negative crystals are reported in Table 2.1

In materials in which phase matching cannot be met it is possible to realise a quasi phase matching (QPM) or **Type 0** phase matching. This consists in the use of a periodically poled crystal in which the direction of the extraordinary axis is inverted every time that the accumulated phase mismatch becomes equal to π . This defines a coherence length given by:

$$L_{coh} = \frac{\pi}{\Delta k}. \quad (2.20)$$

By inverting the axis after each coherence length an additional phase of π is added, so that the phase-matching conditions are set back to the beginning of

the propagation. This results in an increase of the phase-matching efficiency, as pictured in Fig. 2.5.

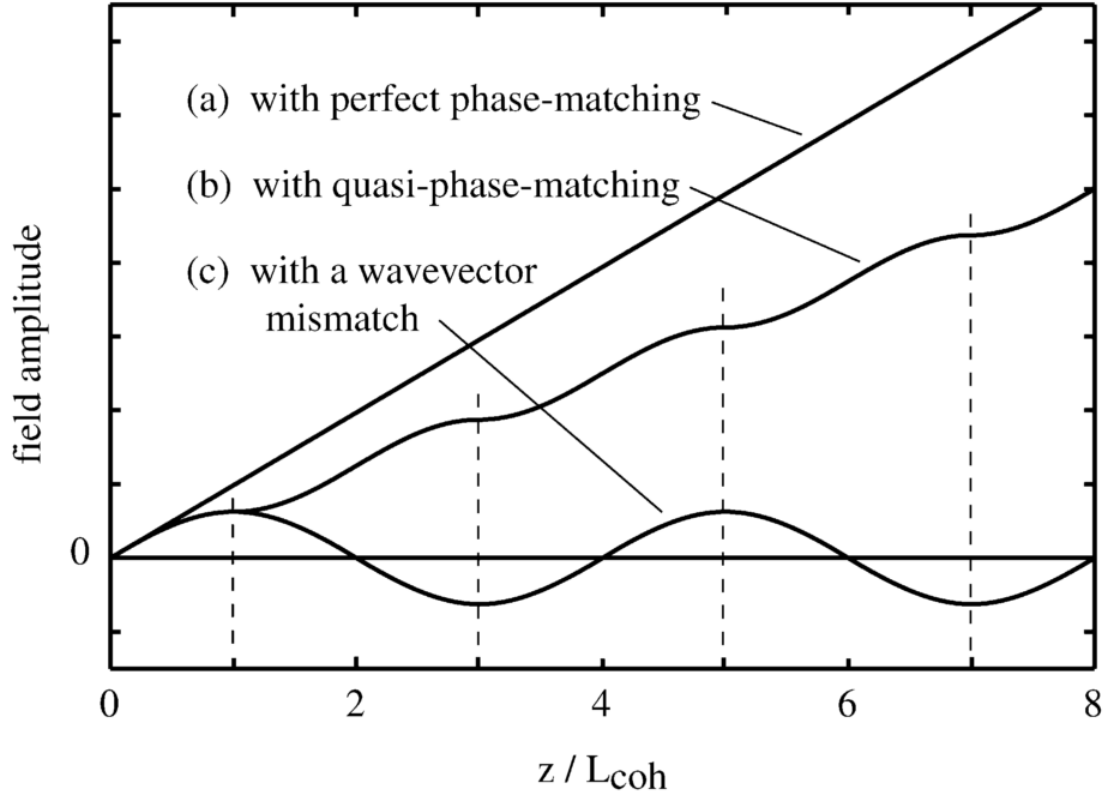


Fig. 2.5 Quasi phase matching [48]. Behaviour of the amplitude of a signal generated in a nonlinear interaction with perfect phase matching (a), quasi phase matching (b), and no phase matching (c) in function of the coherence length.

There is a boundary on how much the beams need to be phase-matched in order for the SFG to happen, in fact the intensity of the outgoing beam is proportional to:

$$I_3 \propto L^2 \text{sinc}^2 \left(\frac{\Delta k(\omega, \theta)L}{2} \right), \quad (2.21)$$

which depends on the phase mismatch Δk and the length of the crystal L . By choosing a suitable L and θ , it is thus possible to model a phase-matching function which can cover a desired bandwidth; in fact, the smaller the length of the crystal, the

larger the bandwidth which can be phasematched, however a thin crystal strongly affect the efficiency so often a trade-off needs to be reached.

2.2.2.1. SELF PHASE MODULATION

Self-Phase Modulation (SPM) is a third-order ($\chi^{(3)}$) process in which the refractive index is modulated by the temporal intensity of the field, so that:

$$n = n_0 + n_2 I(t) \quad (2.22)$$

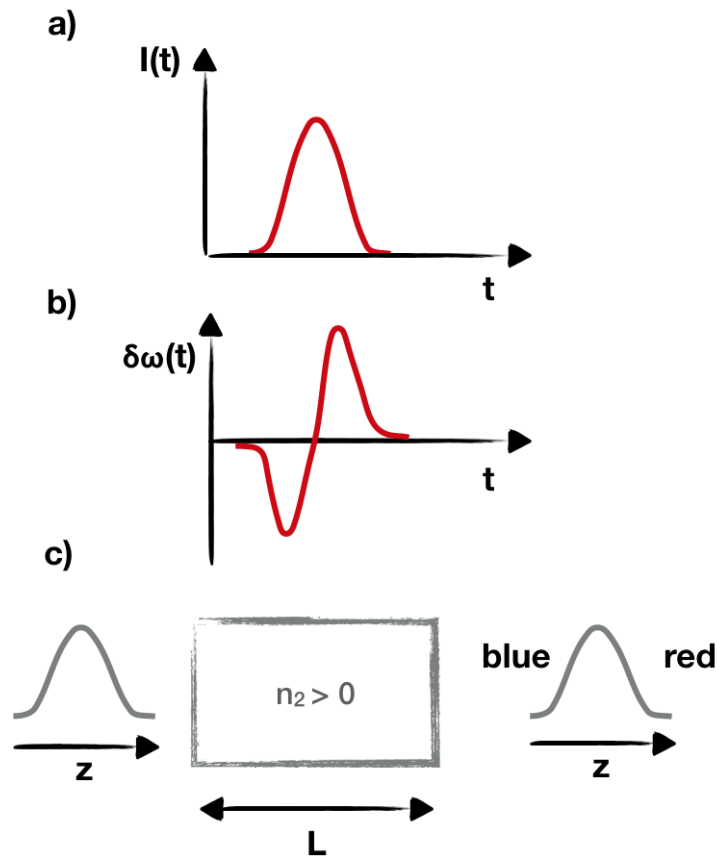


Fig. 2.6 Self Phase Modulation. a) Temporal intensity profile b) frequency shift in function of time c) effect due to the propagation through a medium with $n_2 > 0$

where $n_2 = \frac{\chi^{(3)}}{n_0}$ is the nonlinear refractive index of the medium [48]. The time-dependent refractive index will result in an instantaneous frequency shift $\delta\omega$ which will depend on the derivative of the intensity profile:

$$\delta\omega = -\frac{d\phi_{SPM}(t)}{dt} = -\frac{\omega n_2 L}{c} \frac{dI(t)}{dt} \quad (2.23)$$

where $\phi_{SPM} = -n_2 I(t) \omega L / c$ is the change in the phase of the transmitted pulse due to SPM. As shown in Fig. 2.6 the leading edge of the pulse will be red-shifted while the trailing edge will be blueshifted. SPM is a common method used to obtain spectral broadening [49]. Thus through this nonlinear effect it is possible to generate an output pulse which, if Fourier-limited, is shorter than the input pulse. However, since SPM also introduces a chirp on the pulse, in order to obtain a shorter pulse, compression methods must be implemented after propagation.

2.3. FREQUENCY COMBS

Frequency combs consist of a succession of equally-spaced narrow spectral components. The position of each component is given by:

$$f_n = n \cdot f_{rep} + f_{CEO} \quad (2.24)$$

where f_{rep} is the repetition rate and f_{CEO} is the carrier-envelope offset (CEO) and is given by:

$$f_{CEO} = \frac{\phi_{CEP}}{2\pi} f_{rep} \quad (2.25)$$

The CEO frequency is defined as a constant shift of the carrier-envelope phase between consecutive pulses of a pulse train. In the spectral domain, a CEO frequency

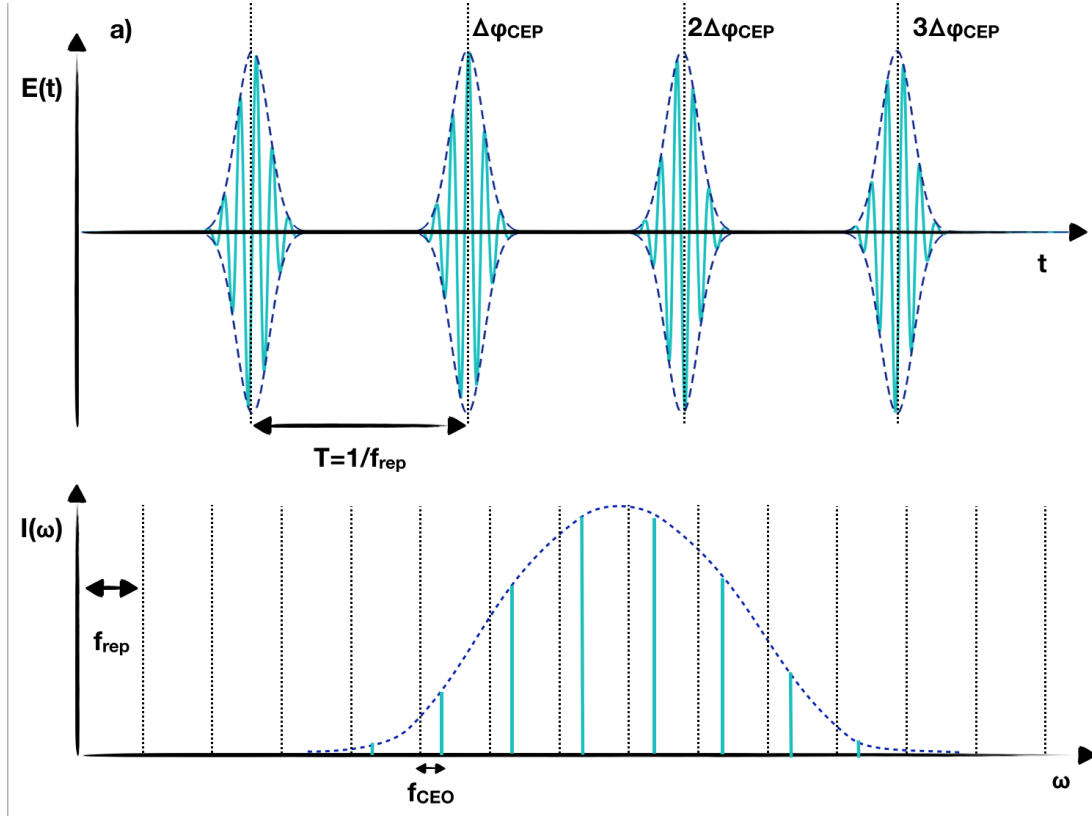


Fig. 2.7 Pulse train and its corresponding frequency comb.

different from zero corresponds to a constant change of the absolute spectral phase (which is related to the CEP). If combining two pulses with different CEO, their absolute phases would change by different amounts resulting in a change of their relative phase.

In Fig. 2.7 a pulse train with a CEO of $\Delta\phi_{CEP}$ in the time domain and the resulting frequency comb is shown.

In order to detect the CEO, the most common method used is an f-to-2f interferometer [50]. This technique requires an octave-spanning bandwidth to work and consists in doubling part of such bandwidth so that it overlaps with the other end of the spectrum, generating beats at the frequency given by f_{CEO} . If the bandwidth is not sufficient, then other methods as f-to-3f interferometry can be used [51].

2.4. LIGHT-WAVE SYNTHESIZERS

In the previous sections it has been shown how the duration of an ultrashort pulse is directly linked to the pulse's spectral bandwidth and the role of dispersion (i.e. the spectral phase), which if correctly controlled allows to achieve a FTL pulse. Having increasingly shorter light pulses is a challenge that the community has been facing in the last few decades to respond to the demands of applications for the study and control of faster dynamical processes and interactions. A first step toward this direction came with the discovery of mode locking [1–3], where single monochromatic cavity modes are coherently combined to obtain a shorter laser emission; Nowadays, few-optical-cycle pulses can be obtained directly from the laser oscillator [7, 8], by spectral broadening within a Hollow-Core Fibre (HCF)[9, 10], by filamentation in gases [11] and by optical parametric amplifiers (OPA) [12]. Although these sources can be all controlled to obtain a FTL pulse, they all have the intrinsic limit set by the period of the carrier wave. However, recently, it has become of paramount importance for many applications (as for example the generation of an isolated attosecond pulse via HHG [29, 30]) to have control over the waveform of ultrashort pulses specifically to obtain pulses shorter than a single optical cycle. To obtain this, very much alike to the modelocking method, it is possible to coherently combine different multi-colored laser sources (parent pulses) so that they will merge to result in a shorter pulse.

For the synthesis to be successful however, the bandwidth and phase of the parent pulses need to satisfy specific requirements; in particular the pulses' bandwidth need to cover a wide spectral range to guarantee enough support for a short pulse. The single parent pulses however can be narrowband and not contiguous, as it is often the case when separated sources are used; this is one of the key motivation

behind this thesis work, as such light synthesizers hence constitute one of the major sources of spectral-gap pulses. The phases of the parent pulses and their relative phase plays a central role in the success of the synthesis:

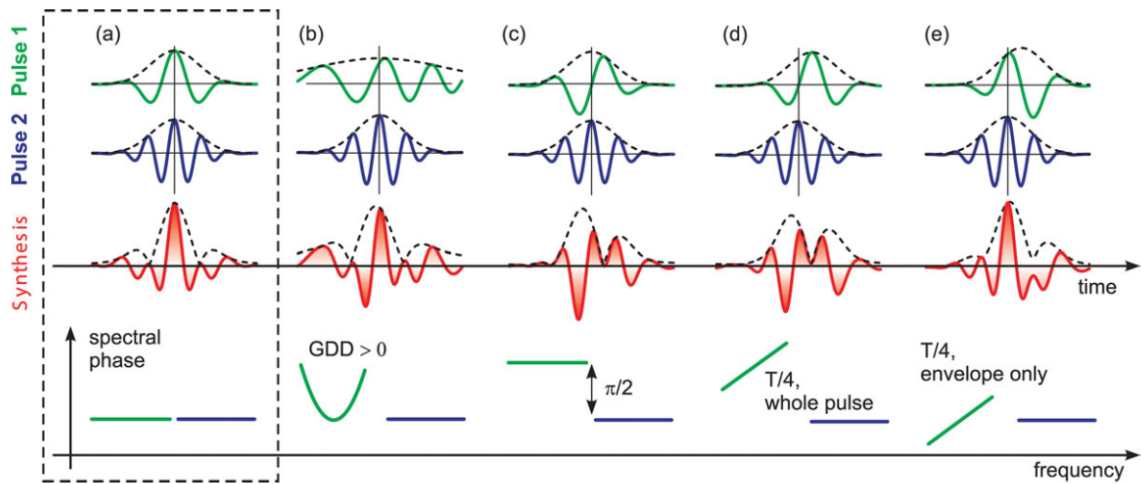


Fig. 2.8 Coherent pulse synthesis [40]. Synthesis of two parent pulses (green and blue) with flat phases and zero relative phase (a), quadratic and flat phases (b), flat phases with $\pi/2$ relative phase (c), linear phase (whole pulse) and flat phase (d), and linear phase (envelope only) and flat phase (e). The synthesised pulse (in red) changes drastically in the scenarios considered; this suggests that a full control of the phases of the parent pulses and their relative phase as well is needed to produce a reliable synthesised pulse.

Fig 2.8 a) shows the ideal case in which both the parent pulses (green and blue) are FTL and have zero relative phase. This configuration leads to the shortest possible synthesis (red). If as in b) one of the pulses were not FTL, but second order phase was added, then the resulting pulse won't be able to reach the same duration as in the FTL scenario. However in c), d) and e) the phases are flat or at most linear and the synthesis is equally affected: this shows that not only dispersion plays a role in the coherent synthesis but also the relative phase (CEP shift) and relative delay between the pulse can drastically change the duration of the resulting pulse. It is straightforward that in order to synthesise such pulses not only the dispersion

but also the CEP and delay need to be controlled. Consequently also measuring such pulses requires a method which grants for the retrieval of the relative phase and delay as well as the dispersion itself.

2.4.1. STATE OF THE ART OF SYNTHESIS SOURCES

In the last decade different approaches have been undertaken to obtain tailored synthesized pulses.

In [52], Krauss et al. use a 40 MHz erbium-doped fiber oscillator to obtain a broadband pulse centered at 1125 nm and a soliton at 1700 nm, resulting in a 4.3 fs single-cycle waveform starting from 7.8 fs and 31 fs parent pulses (Fig. 2.9 a)).

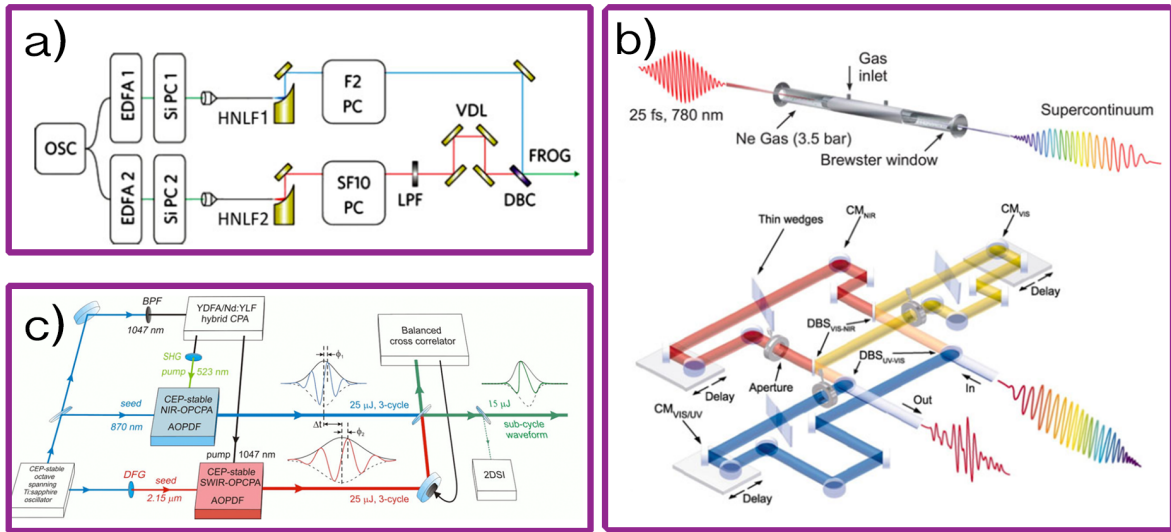


Fig. 2.9 Experimental setups from [40]. (a) Scheme of a fiber-based single-cycle waveform synthesizer. OSC: femtosecond erbium-doped fiber oscillator; EDFA: erbium-doped fiber amplifier; Si PC: silicon prism compressor; HNL F: bulk highly nonlinear fiber; F2/SF10 PC: pulse compressors using F2 and SF10 Brewster prisms; LPF: low-pass filter VDL: variable delay line; DBC: dichroic beam combiner. (b) Supercontinuum generation in a neon-filled hollow-core fiber and schematic representation of a three-channel light-field synthesizer. (c) Scheme of the high-energy OPCPA waveform synthesizer. BPF: bandpass filter; SHG: second-harmonic generation crystal; DFG: intrapulse difference-frequency generation module; AOPDF: acousto-optic programmable dispersive filter

In [27] Wirth et al. use as parent pulses slices of the output of an HCF filled with neon and seeded with a Ti:Saph laser. The three parent pulses so obtained are individually compressed and coherently combined to obtain a sub-cycle waveform with a 2.1 fs pulse width (Fig. 2.9 b)).

In [53] Huang et al. demonstrate an OPCPA-based pulse synthesizer,(Fig. 2.9 c)), by coherently combining two OPCPA one amplifying a signal at 870 nm and the other at 2150 nm. Each of the OPCPA has also an acousto-optic programmable dispersive filter (AOPDF) which allows the individual control of amplitude and spectral phase. The outputs of the OPCPA are both 3-cycle pulses (with a duration of 9 fs and 24 fs for the 870 nm and 2150 nm respectively) which combine into a sub-cycle waveform, lasting 0.8 cycles of the carrier frequency of 1260 nm.

Another attempt of coherent pulse synthesis which will be discussed more in detail in Chapter 4 is [54] where McCracken et al. propose a scheme aiming at obtaining optical attosecond pulses (OAP) in the visible regime ⁴. Using an OPO the bandwidth required is covered by upconverting an incoming pump beam by means of a tailored nonlinear crystal so that different combinations of pump, signal, and idler can be obtained at the same time. All the sources here mentioned, apart from that in [27] exhibit separated spectral components.

⁴ To actually obtain an attosecond duration the visible bandwidth is not enough, there is need to use also light in the near UV. This however is opposed to the XUV regime where attosecond pulses are commonly obtained by HHG

ULTRAFAST METROLOGY

Five hundred twentyfive thousand six hundred minutes
How do you measure, measure a year?
In daylights, in sunsets, in midnights, in cups of coffee
In inches, in miles, in laughter, in strife
In, five hundred twentyfive thousand six hundred minutes
How do you measure a year in the life.

Rent, Jonathan Larson

As the quote at the beginning of this chapter states, there are different ways in which a time interval can be measured: depending on the time scale and the properties we want to underline we might use different methods, but what they all have in common is that they make use of something with a shorter duration than the time interval itself. So what happens when the duration we want to measure is also the shortest we can achieve? This has been one of the main problems in the laser community since mode locked lasers [2, 3] provided for the first time a pulse with a bandwidth large enough to obtain a FTL duration shorter than any photodetector response time.

When dealing with pulses on the order of the picosecond or longer, the time duration can be directly measured by means of a streak camera or a photodiode. If also the spectrum is provided, it is possible to obtain information on the time bandwidth product. By comparing it with the time duration obtained, one can draw considerations on the presence of a varying spectral phase. If the pulses are shorter, however, this is not possible as the response of the detectors is longer than the pulses; thus to determine their time duration, indirect measurements need to be implemented. As we have discussed in the previous chapter the pulses can be described by a complex field, thus they are uniquely determined either in the spectral or temporal domain by an amplitude and a phase. Measuring the amplitude is an easy task which can be performed with any spectrometer, but gathering information on the phase instead has posed many challenges and many techniques have been developed to provide a solution to this longstanding problem. Most instances have been successfully addressed, but some more complex cases remain elusive with state-of-the-art techniques.

The first attempts at determining the duration of an ultrashort pulse have been based on the notion that since there weren't any shorter known references, the pulse needed to act as a reference itself and information can be extracted via its autocorrelation [55, 56]. Two main autocorrelation techniques were developed and are schematically pictured in Fig. 3.1: intensity and interferometric autocorrelation.

In the intensity autocorrelation scheme, the pulse is split via a beam splitter and the two resulting fields are overlapped on a nonlinear crystal as their relative time delay is scanned. The second harmonic signal generated on the nonlinear crystal is recorded with a photodetector in function of the time delay. Since the signal is non-zero only when the two fields are simultaneously present on the crystal, by analyzing the upconverted signal it is possible to extract information on the time

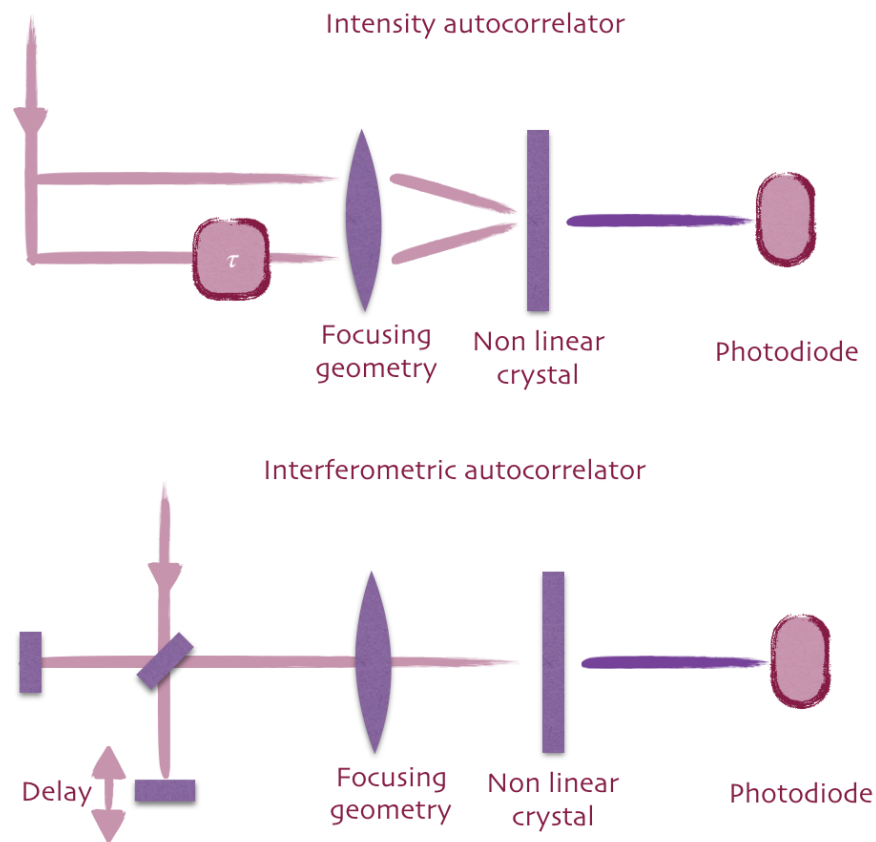


Fig. 3.1 Autocorrelator schemes. Intensity autocorrelator: the beam is split into two arms; a delay is applied to one of the arms and they are both focused on a nonlinear crystal resulting in a SHG signal. The signal is then recorded with a photodiode. Interferometric autocorrelator: The beam is sent on a Michelson interferometry and the output is focused on a nonlinear crystal obtaining a frequency-doubled signal which is then recorded with a photodiode.

duration of the original pulse, however without obtaining any information on the phase. In the interferometric autocorrelation scheme the pulse enters a Michelson interferometer with an adjustable arm to scan the relative time delay; one of the two output of the interferometer is sent through a nonlinear crystal and then recorded with a photodetector. As the resulting signal (in Fig. 3.2) contains products of the electric field, it also bears some phase information, which however can only be successfully extracted in specific cases [57]. A qualitative conclusion on whether the

pulse is FTL or chirped can be quickly obtained from the trace as for chirped pulses the fringes will not appear on the wings (Fig 3.2 b).

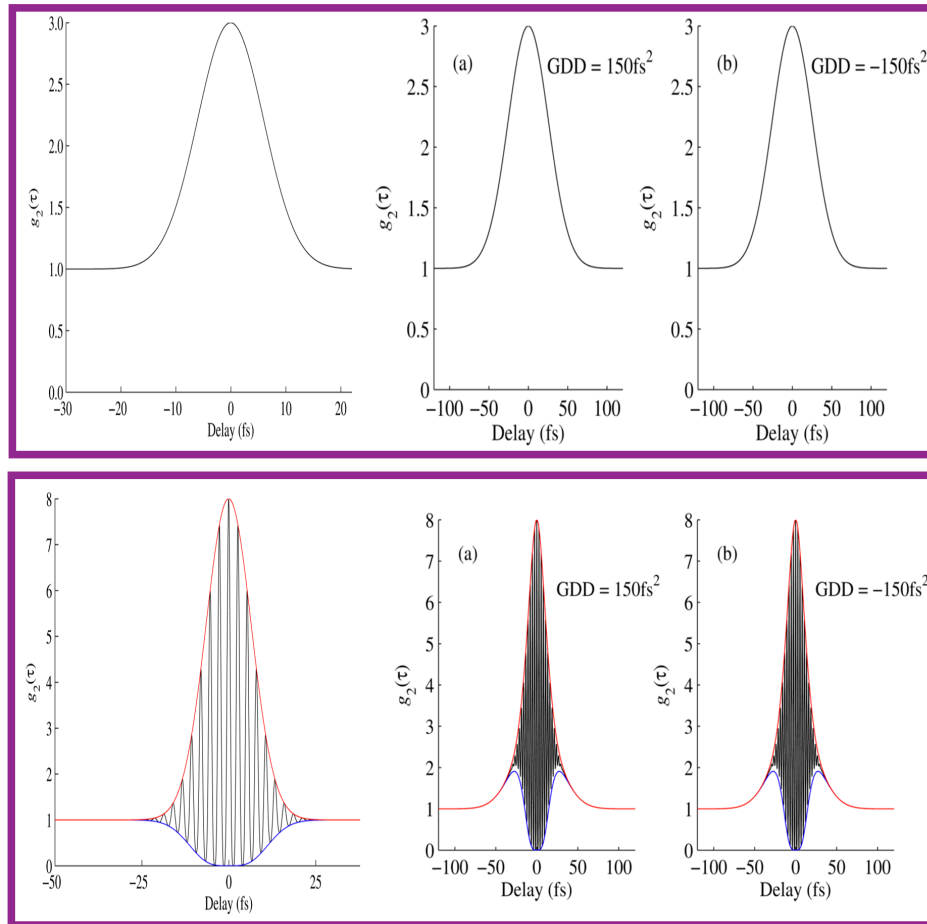


Fig. 3.2 Intensity (upper panel) and Interferometric (lower panel) autocorrelation signal for a FTL pulse (left), a pulse chirped with $GDD=150 \text{ fs}^2/\text{rad}$ (centre), and a pulse chirped with $GDD= -150 \text{ fs}^2/\text{rad}$ (right).

While some information on the temporal duration of the pulses can be extracted via autocorrelation, to obtain a full phase characterisation more complex techniques need to be implemented. Several techniques have been developed to retrieve both the intensity and phase in the last few decades and can be divided in two categories: *spectrographic* techniques, based on the autocorrelation methods just discussed, and *interferometric* techniques which rely on direct phase extraction from interference

signals. Each category presents many different variants which can be chosen to suit best the experimental needs case by case. This chapter will provide an overview on the most used techniques with particular attention to those that will pave the way for the newly developed technique analysed in Chapter 5. Furthermore a new phase retrieval algorithm (the MICE algorithm) is discussed, and the topical problem of retrieving the phase with spectral nulls, which is the core problem of this work, is introduced with particular emphasis on the state of the art and limitations of the different existing techniques. We shall also see how this is relevant to the investigation of advanced light sources.



3.1. SPECTROGRAPHY

Spectrographic techniques have been among the first attempts to characterize optical pulses [58–60]. The concept behind these range of techniques is to retrieve the information about the pulse which is captured as a joint representation of the Fourier conjugate variables time and frequency. While these concepts seem unintuitive, they actually represent the way a musical score is built, hence this is a proper example of a simple spectrographic measurement:



Fig. 3.3 Spectrogram of a musical (excerpt from Les Misérables, by Boublil & Schoenberg).

In Fig. 3.3 the notes are in fact disposed on the pentagram according to their frequency which is discrete and set by the well tempered scale (an octave is divided into 12 equal intervals on a logarithmic scale [61]), and they succeed one after the other in a temporal fashion set by the time signature displayed after the key. In the same way as notes are written on a musical score, in a *spectrogram* the spectrum of the signal is recorded as a function of the time delay with respect to the pulse peak. Mathematically a spectrogram is defined by the function:

$$S(\omega, \tau) = \left| \int_{-\infty}^{\infty} E(t) g(t - \tau) e^{(-i\omega t)} dt \right|^2 \quad (3.1)$$

where $E(t)$ is the electric field and $g(t - \tau)$ is known as the gating function which selects sections of $E(t)$ as the delay τ varies [62]. The probe and gate functions can be either obtained from the same identical pulse, in which case the method

3.1 Spectrography



would be self-referencing, or from different pulses in which case a known reference can be used.

Although the spectrogram is an intensity measurement and thus does not provide any direct information on the phase, it is enough in most cases to fully characterize the field $E(t)$. There can however be ambiguities which we will discuss later in the chapter.

3.1.1. FROG

The most common example of spectrographic technique is the Frequency Resolved Optical Gating [14, 58, 63]. FROG is an autocorrelation-like measurement in which the upconverted signal from an intensity autocorrelator is spectrally resolved and recorded as a function of the time delay between the test pulse and a gate pulse. This results in a $N \times N$ points trace, where N is the sampling size, from which $2N$ points (intensity and phase of the test pulse) are retrieved by means of an iterative algorithm. There are different types of FROG implementations which differ from the geometry and the gating function used, each tailored to specific measurement requirements [14, 63–66]. Here I describe three of the most common variants, SHG, PG, and X FROG.

3.1.1.1. SHG FROG

The most widely used FROG configuration is Second Harmonic Generation (SHG) FROG [65, 67–71]. The SHG FROG scheme is shown in Fig. 3.4: the test pulse is mixed with its time-delayed replica on a nonlinear crystal and the resulting second harmonic signal is recorded with a spectrometer as a function of the delay between the replicas.

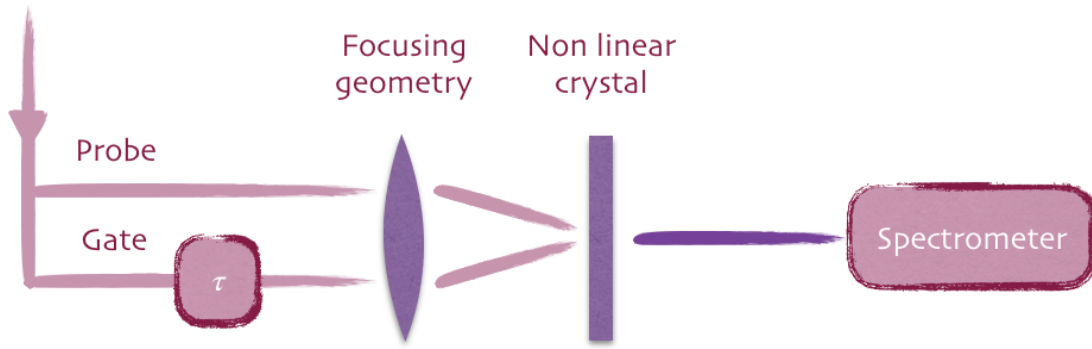


Fig. 3.4 Example of a SHG FROG setup: in the same fashion that with an intensity autocorrelator, the beam is split into two arms, one of which undergoes through a delay line. Both beams are focused on a nonlinear crystal where they generate a SHG signal, which is then recorded with a spectrometer in function of the time delay.

The SHG FROG trace is given by:

$$S_{SHG}(\omega, \tau) = \left| \int_{-\infty}^{\infty} E(t)E(t - \tau)e^{-i\omega t} dt \right|^2, \quad (3.2)$$

of which some examples are given in Fig. 3.5. The main disadvantage of SHG-FROG is that, contrary to other geometries, the traces are unintuitive and show an ambiguity in the time direction: considering a test pulse $E(t)$ or $E(-t)$ would lead to the same trace. This ambiguity is however trivial and can be removed by performing also a measurement inserting a known amount of dispersion in the beam before it is split. However this easy fix is not favorable when a single-shot measurement is required. An alternative solution has been proposed by Taft [72], which consists of inserting an etalon before the beam splitter so that the back reflection provides a trailing (or leading) satellite pulse depending on the time direction. SHG FROG is however widely used as it utilizes a very standard setup and, since the signal is obtained with a second order nonlinearity, it is advantageous with weak pulses

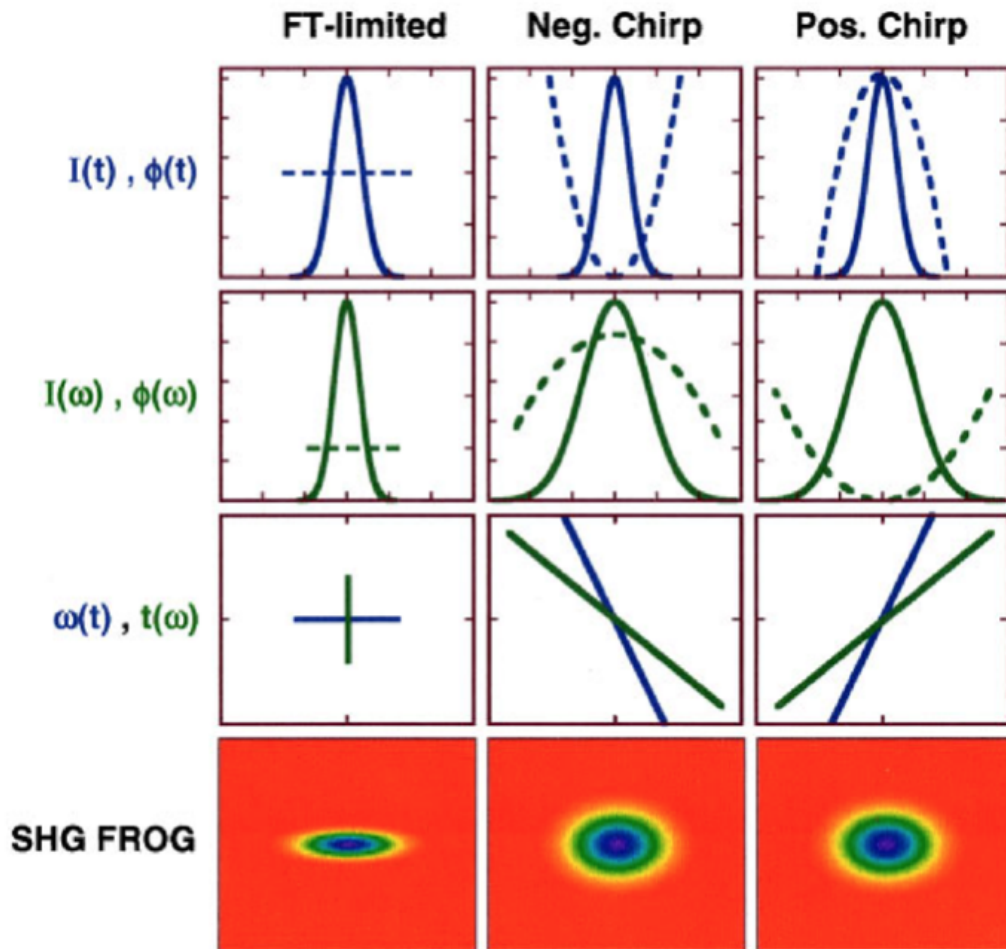


Fig. 3.5 Example of SHG FROG traces from [63]. First row: intensity and phase in the time domain; second row: intensity and phase in the frequency domain; third row: instantaneous frequency vs. time and group delay vs. frequency. forth row: SHG FROG trace. These are shown in the instance in which the pulse is FTL (left column) or has a negative (centre column) or positive (right column) chirp.

compared to other geometries which adopt a third order process for the signal's generation.

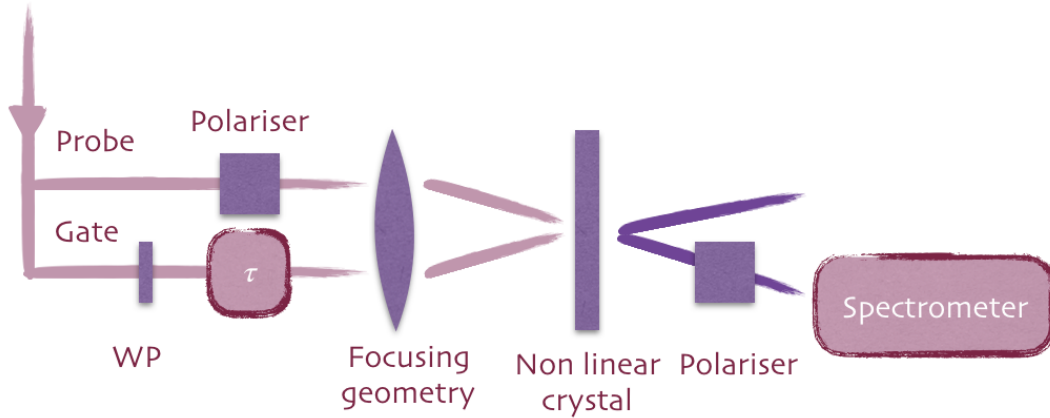


Fig. 3.6 PG-FROG setup scheme: the probe pulse is sent through a polarizer and the gate pulse through a HWP which rotates its polarisation by $\pm 45^\circ$ and is then delayed via a delay line. Both beams then overlap into a third-order nonlinear medium. The signal obtained after the interaction on the nonlinear crystal will thus be polarisation-dependent and will be selected with a polarizer before being measured with a spectrometer in function of the time delay.

3.1.1.2. PG-FROG

Polarisation Gate (PG) FROG [58, 60, 73, 74] utilizes a slightly more demanding setup compared to SHG FROG, which allows to gate the pulse by means of polarisation analysis. The PG-FROG scheme is shown in Fig. 3.6: the probe pulse is sent through a polarizer and the gate pulse through a HWP which rotates its polarisation by $\pm 45^\circ$ and is then delayed via a delay line. Both beams then overlap into a third-order nonlinear medium. In the medium the gate introduces a birefringence due to the electric Kerr effect, which causes the probe pulse polarisation to be slightly rotated for the extent of the interaction. The signal is then recorded after a polarisation analyzer to select only the rotated polarisation light, which is only present due to the gate pulse. The trace reads:

$$S_{PG}(\omega, \tau) = \left| \int_{-\infty}^{\infty} E(t) |E(t - \tau)|^2 e^{-i\omega t} dt \right|^2. \quad (3.3)$$

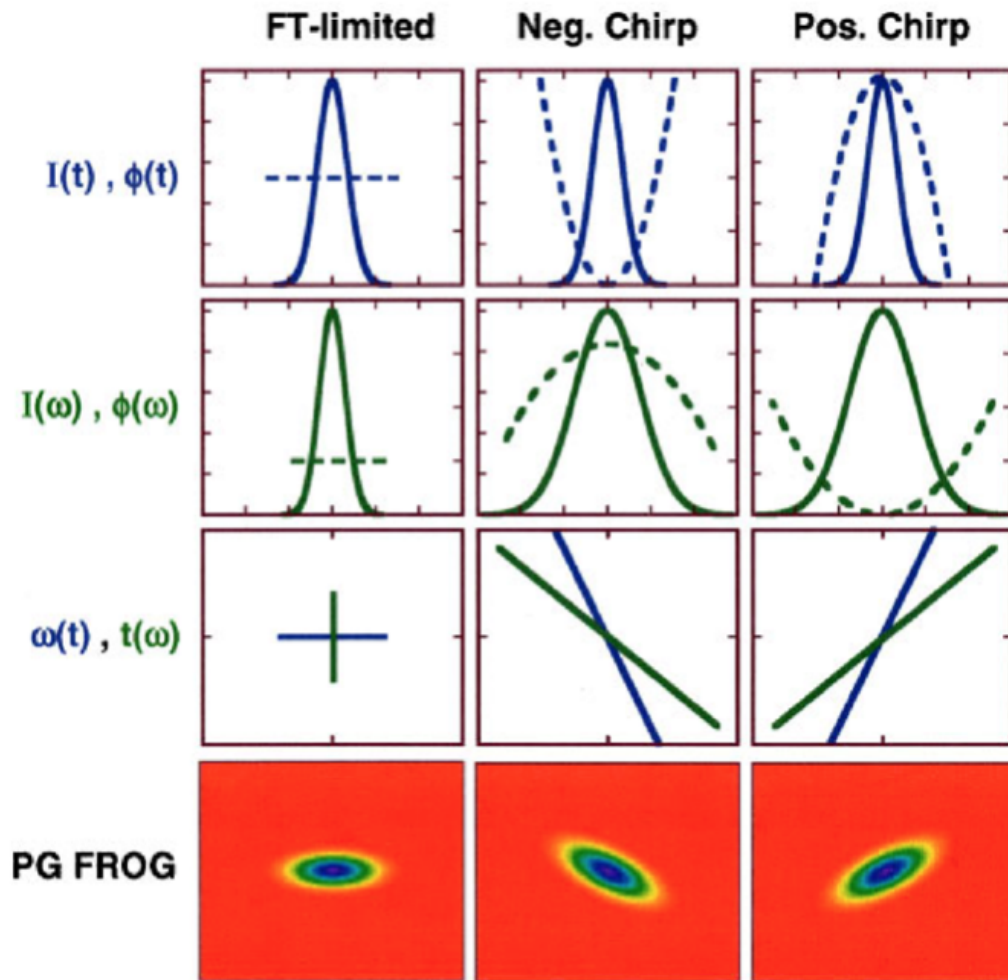


Fig. 3.7 Example of PG-FROG traces from [63]. First row: intensity and phase in the time domain; second row: intensity and phase in the frequency domain; third row: instantaneous frequency vs. time and group delay vs. frequency. forth row: PG-FROG trace. These are shown in the instance in which the pulse is FTL (left column) or has a negative (centre column) or positive (right column) chirp.

The traces, of which an example is given in Fig. 3.7 appear much more intuitive than in the SHG configuration and that is because in (3.3) the gate appears as a real quantity thus adds no phase information to the gated slice of the probe pulse. Another advantage of PG-FROG is that it is not affected by any trivial ambiguities as instead it is the case with SHG FROG. This is however compensated by a much



more expensive setup as to be working properly high-quality polarizers must be used and, due to the third-order nonlinearity it is not suited for low-energy pulses.

3.1.1.3. X FROG

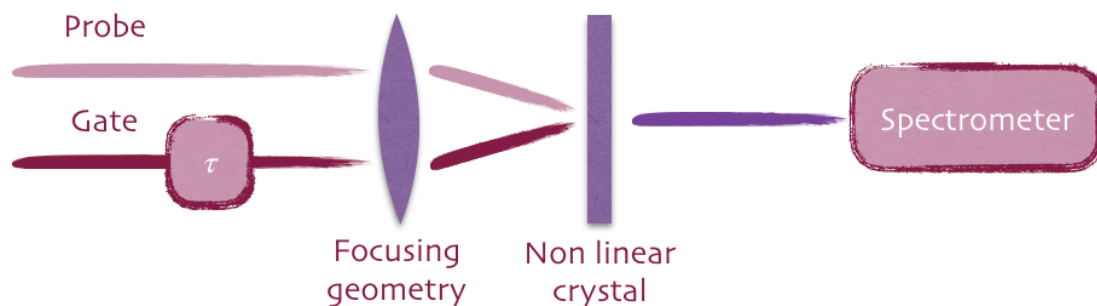


Fig. 3.8 X-FROG setup scheme: two different beams (probe and gate), one of which undergoes through a delay line, are focused on a nonlinear crystal. A SHG signal is generated and recorded by means of a spectrometer.

X FROG [16] is the cross-referenced configuration of the FROG technique and allows for the characterisation of even weaker pulses than the SHG variant. As well as SHG FROG, X FROG utilises a second order nonlinearity to obtain the signal pulse (sum frequency generation), but with the additional freedom of having a powerful reference acting as a gate to compensate for the weak probe pulse. The downside is that the reference needs to be well characterized in advance, and tailored to the particular probe pulse considered.



3.1 Spectrography

The experimental scheme of X FROG is shown in Fig. 3.8, and apart from the use of an independent reference, is identical to the SHG configuration. The trace resulting from an X FROG measurement is given by:

$$S_{XFROG}(\omega, \tau) = \left| \int_{-\infty}^{\infty} E_{Probe}(t) E_{REF}(t - \tau) e^{-i\omega t} dt \right|^2 = \left| \int_{-\infty}^{\infty} E_{Cross}(t, \tau) e^{-i\omega t} dt \right|^2. \quad (3.4)$$

Despite being equivalent to the SHG configuration, X FROG is not affected by the same ambiguity because the two pulses are not identical, thus the traces result more intuitive than the SHG ones.

3.1.1.4. PHASE RETRIEVAL ALGORITHM

FROG uses an iterative algorithm to extract the spectral phase from a spectrogram, the principal component generalised projections algorithm (PCGPA) [75]. This algorithm is used to reconstruct the two fields that contribute to the FROG trace which is a real quantity and thus does not contain directly any phase information. In principle the two fields can be different, which is the case of blindFROG [75] but as it is discussed later in the chapter, if the gate and probe pulses are not the same, many ambiguities arise which are instead confined when the two pulses are identical.

The aim of the PCGPA algorithm is to determine two fields whose outer product matches the measured FROG trace within a defined error threshold. The procedure is shown in Fig. 3.9: the two fields are initialized with a random guess of N points sampled with the time delay τ ; an outer product O of the two vectors is obtained, and in order to get the delay on the gate pulses as of (3.1), a row rotation is performed on O before Fourier transforming and applying a constrain on the transformed outer product. In the PCGPA, the Fourier domain constraint consists in replacing the

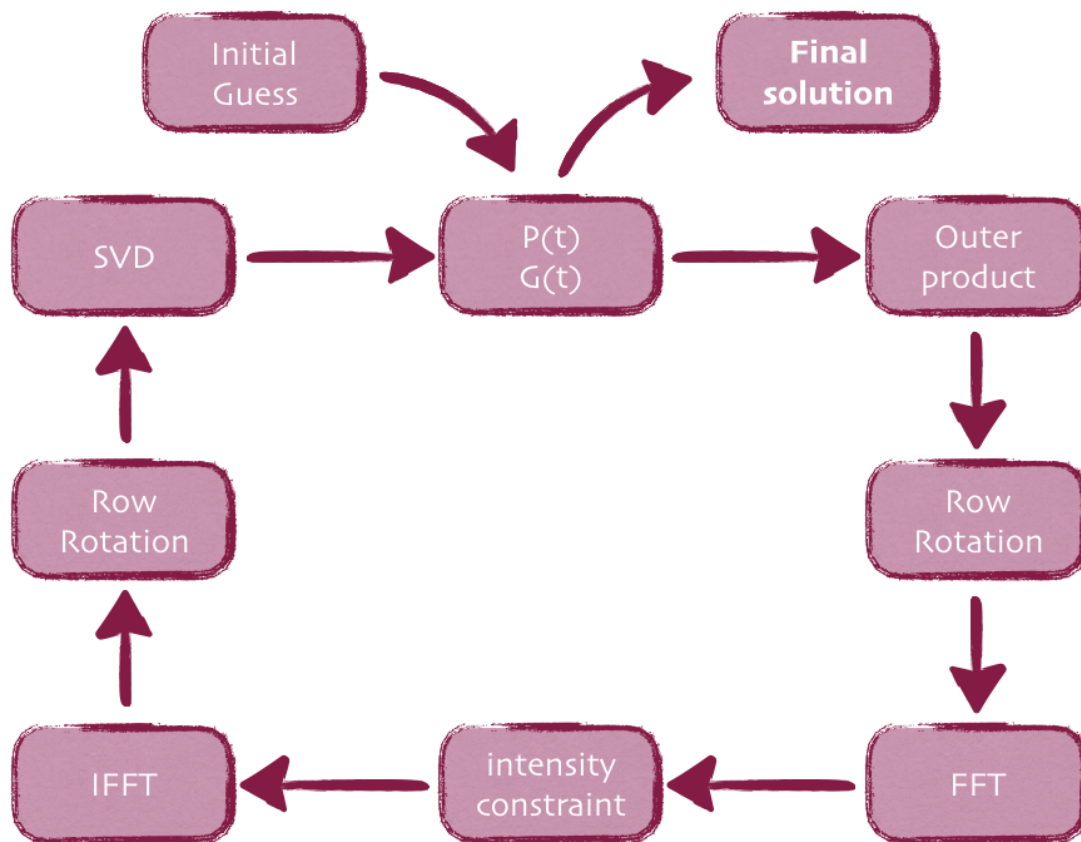


Fig. 3.9 FROG algorithm scheme

magnitude of the new trace by the experimentally measured trace. The so-obtained new trace is then reverted back to the time domain with an inverse Fourier transform and the time delay is taken off from the gate with a row rotation. The two new fields are extracted by means of a procedure called singular value decomposition (SVD) [76]. SVD decomposes a matrix into a superposition of outer products of vectors. Only the outer product pair with the principal contribution is kept and utilized as the

3.1 Spectrography



new guess for the two fields in the next iteration. The procedure is repeated until the FROG error

$$\epsilon = \left[\frac{1}{N^2} \sum_{i,j=1}^N [I_{recon}(\omega_i, \tau_j) - I_{meas}(\omega_i, \tau_j)]^2 \right]^{1/2}, \quad (3.5)$$

where I_{recon} is the reconstructed intensity and I_{meas} is the measured one,

reaches the desired value. Even if the convergence is not assured [77, 78], when dealing with the same identical field for the probe and the gate the algorithm is very robust and not subject to nontrivial ambiguities.

The iterative retrieval algorithm for the referenced configuration is similar to the one just described but utilises an additional constraint, as illustrated in Fig. 3.10: a random initial guess for $E_{Probe}(t)$ is used together with the known reference field to obtain the Fourier transform of $E_{Cross}(t, \tau)$, $E_{Cross}(\omega, \tau)$. This is then fitted to the X FROG trace by replacing its magnitude with the square root of S_{XFROG} , obtaining $E'_{Cross}(\omega, \tau)$ which is then Fourier transformed back to $E'_{Cross}(t, \tau)$. The obtained product is used to obtain a new guess for $E_{Probe}(t)$ with the method of generalized projection [77], by minimizing the metric:

$$Z = \sum_{i,j=1}^N |E'_{cross}(t_i, \tau_j) - E_{Probe}(t_i)E_{Ref}(t_i - \tau_j)|^2 \quad (3.6)$$

with respect to $E_{Probe}(t)$.

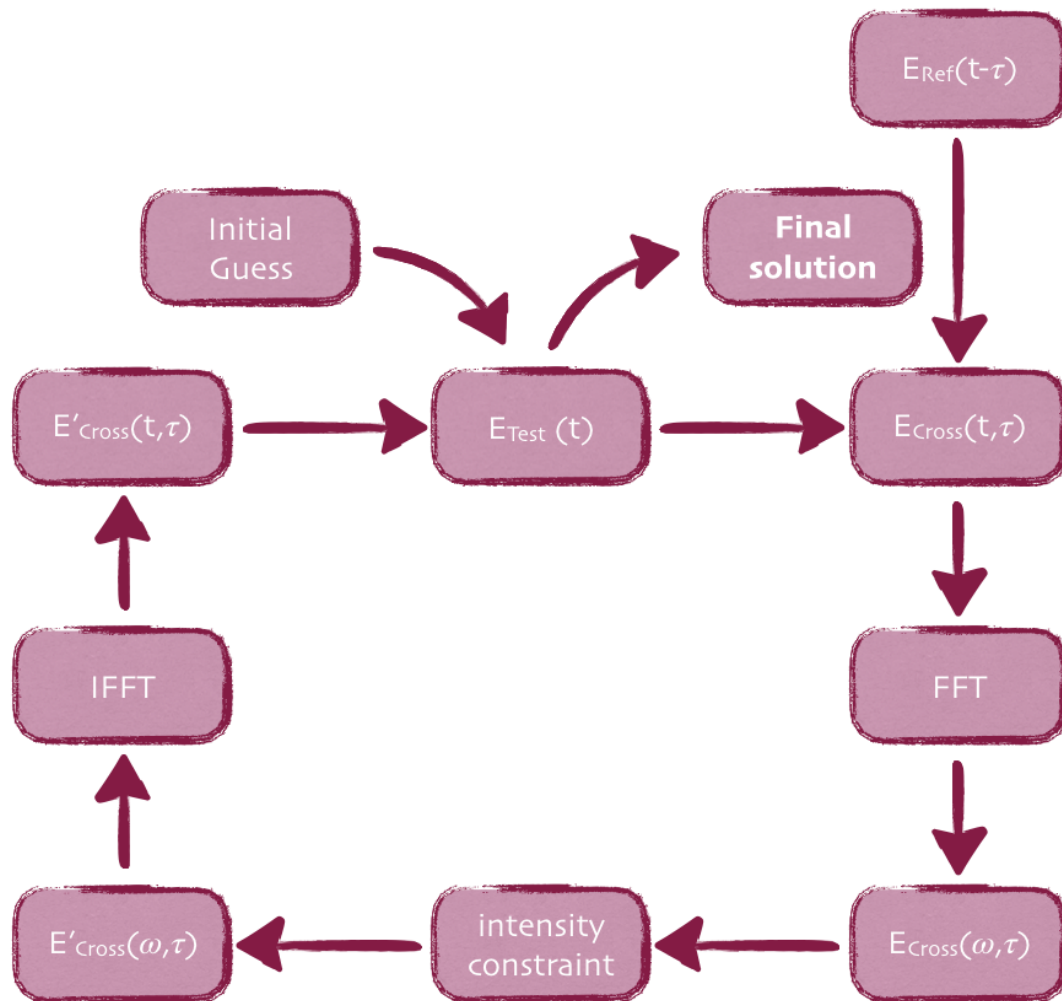


Fig. 3.10 X-FROG algorithm scheme

3.2. INTERFEROMETRY

Interferometric techniques are another widely spread method for measuring ultra-short pulses. The core of these techniques is the extraction of the phase information stored in the fringe pattern given by the interference between two pulses. The concept has been developed starting from the scenario in which one unknown test pulse is interfered with a reference pulse with well-known phase. Delaying the

reference in time will result in an added linear spectral phase term $e^{\pm i\omega\tau}$, where τ is the time delay imposed; so if $E_i(\omega) = |E(\omega)|e^{i\phi(\omega)}$ and we label 1 and 2 the test and reference pulses, then the interferogram will read:

$$I(\omega) = |E_1(\omega)|^2 + |E_2(\omega)|^2 + 2|E_1(\omega)||E_2(\omega)| \cos(\phi_1(\omega) - \phi_2(\omega) + \omega\tau). \quad (3.7)$$

The interference given by the cosinusoidal term stores the information about the phase difference between the test and reference pulses. Provided the reference is known, the unknown phase can be extracted with an adequate algorithm.

Having a reference is a huge constraint for a metrology technique especially as the reference needs to be tailored to the particular test pulse making it less adaptable and scalable to a general case. However from Eq. 3.7 if the reference were to be replaced with an identical copy of the test pulse, the whole phase information would be lost. A further step is then needed to implement a self-referenced technique and the result goes under the name of "Spectral Shearing Interferometry" (SSI) [79–81]. If in fact we take a frequency-shifted copy of the test pulse instead, $E(\omega - \Omega)$, where Ω is referred to as the *spectral shear*, the interferogram of Eq. 3.7 becomes

$$I(\omega) = |E_1(\omega)|^2 + |E_1(\omega - \Omega)|^2 + 2|E_1(\omega)||E_1(\omega - \Omega)| \cos(\phi_1(\omega) - \phi_1(\omega - \Omega) + \omega\tau). \quad (3.8)$$

Thus in SSI the interference pattern provides information about the relative phase between two spectral components of the test pulse separated by the shear Ω . This allows for the complete reconstruction of the phase with a resolution imposed by the shear, which has to comply with the Whittaker-Shannon sampling theorem. For a pulse with compact temporal support, T , the spectrum needs to be sampled with a

maximum angular frequency $2\pi/T$,

$$\Omega \leq \frac{2\pi}{T}. \quad (3.9)$$

At the same time however choosing a shear which is too small compared to the pulse phase features results in a higher sensitivity to noise and shot-to-shot instabilities.

Equivalently to introducing a spectral shear, it is also possible to introduce a temporal shear and performing temporally resolved measurements; however, when dealing with sub-picoseconds pulses the detectors are too slow for this type of measurements thus the frequency domain is preferred [13].

3.2.1. SPIDER

SPIDER (*Spectral Phase Interferometry for Direct Electric-field Reconstruction*) [13, 82] is a particular type of SSI where the shear between the two time-delayed replicas is obtained by means of a nonlinear process. This is made possible by the use of a highly-chirped ancillary beam: consider a pulse given by $E(\omega) = |E(\omega)|e^{i\phi(\omega)}$ and its identical time-delayed copy which can be obtained by taking the front and back reflection of an etalon; the two replicas of the test pulse are then focused in a BBO crystal where they are upconverted with a chirped ancillary pulse derived from the same original pulse. Since the two replicas are delayed in time, they will upconvert with two different quasi-monochromatic frequencies from the chirped pulse. This will generate two signal pulses (SP) sheared in frequency by an amount Ω which is related to the time delay and to the group delay dispersion (GDD) of the chirped pulse by:

$$\Omega = -\frac{\tau}{GDD} \quad (3.10)$$

The resulting interferogram following to the superposition of these frequency sheared pulses is:

$$I_{spider}(\omega) = |E(\omega)|^2 + |E(\omega - \Omega)|^2 + 2|E(\omega)||E(\omega - \Omega)| \cos(\phi(\omega) - \phi(\omega - \Omega) + \omega\tau). \quad (3.11)$$

The oscillatory term containing the information on the phase, is constituted by a carrier $\omega\tau$, which results in spectral fringes with spacing of $2\pi/\tau$, modulated by the difference between the spectral phases of the two replicas.

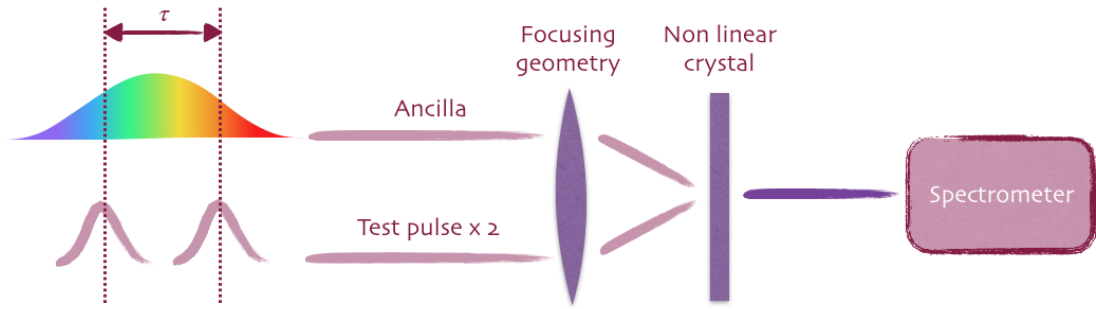


Fig. 3.11 SPIDER scheme. Two identical replicas of the test pulse are focused on a nonlinear crystal where they are upconverted with two monochromatic slices of a temporally chirped ancilla. The interference pattern between the two so-generated signals is recorded by means of a spectrometer.

The phase information contained in $\phi(\omega)$ can be extracted via the reconstruction process described in Fig. 3.11 based on Fourier filtering (Takeda algorithm [83]). If the delay is large enough, when Fourier transforming the spectrum it is possible to isolate one of the two sidebands by amplitude filtering. Then performing a the inverse F.T. on the filtered sideband allows to extract the phase difference:

$$\theta(\omega) = \phi(\omega) - \phi(\omega - \Omega) + \omega\tau \quad (3.12)$$

The $\omega\tau$ term can thus be removed with a calibration step consisting in taking a measurement at $\Omega = 0$.

In order to extract the spectral phase $\phi(\omega)$ from the obtained phase difference, a further step is needed. This can be performed in two different ways, via integration or via concatenation [84]. This step is important because it allows the understanding of two important features of SPIDER: once the phase for a given frequency ω_k is arbitrarily set, the phase on the next sampling point, $\omega_k + \Omega$ can be calculated as:

$$\phi(\omega_k + \Omega) = \phi(\omega_k) + \theta(\omega_k + \Omega) \quad (3.13)$$

This is the reason why SPIDER, as with all the other self-referencing techniques, is not able to retrieve the absolute spectral phase of a pulse: the phase of the first frequency sampling point must be arbitrarily chosen so there is no information about the absolute phase. The other important thing to consider is that if the phase information at a given frequency is lost, for example because the intensity of the signal pulse in that point is zero, then the concatenation fails and there is need to set a new arbitrary phase to reconstruct the phase after that point.

The advantages of the SPIDER technique are many: especially in the implementation just described. The setup is simple and combined with the fast algorithm makes SPIDER an ideal candidate for online routine diagnostic, also favoured by the single-shot capabilities of this technique. Furthermore the measurements are very robust to noise [85].

During the last decade many different variant of SPIDER have been realized to improve and adapt the technique to satisfy the different needs dictated by the pulse complexity [18, 19, 86–95]; . Here I will give a brief description of those three versions of SPIDER will be considered later in this work.

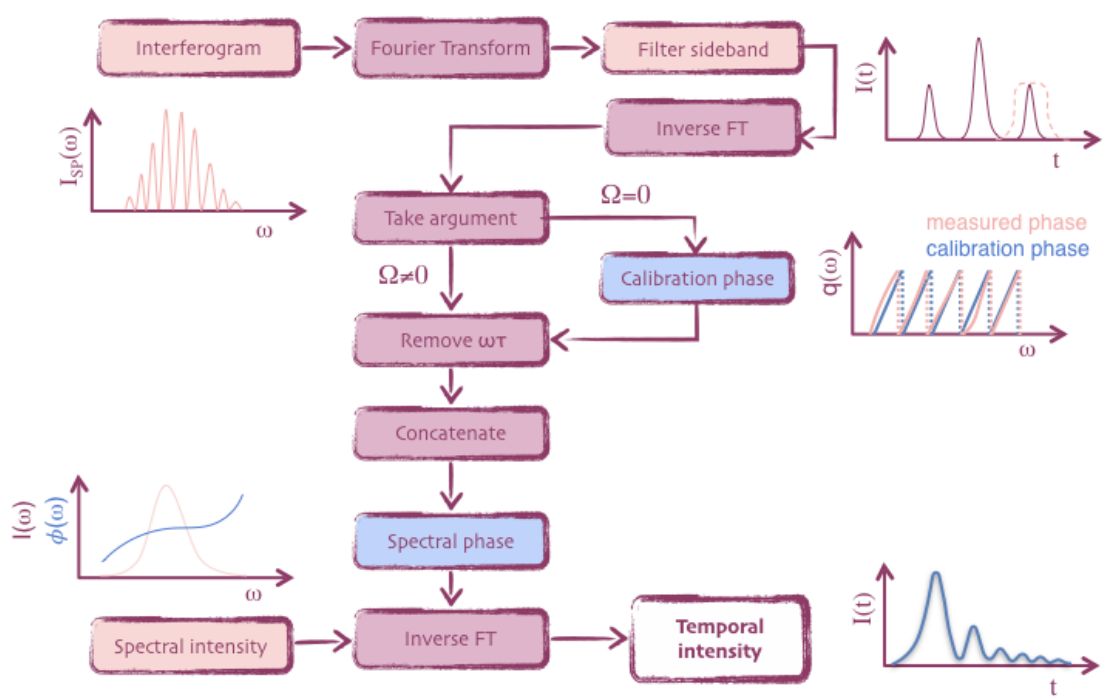


Fig. 3.12 SPIDER phase retrieval scheme.

3.2.1.1. X SPIDER

Exactly as with FROG, a common variant of SPIDER is its cross version, X SPIDER [86, 87, 94, 95]. In X SPIDER the ancillary chirped pulse is not derived from the same pulse as the SP, but can be taken from a more powerful beam which can be useful to improve the reconstruction and the signal-to-noise ratio when dealing with weak test pulses. Furthermore it can be also useful in the instances where frequency doubling cannot be obtained and is preferable to attain frequencies to match the detector.

3.2.1.2. SEA-SPIDER

Spatially Encoded Arrangement-SPIDER (SEA-SPIDER) [13, 18, 21, 93] is a technique that makes use of the spatial domain to enable the signal to be sampled at the minimum spectral sampling rate satisfying the WS sampling theorem.

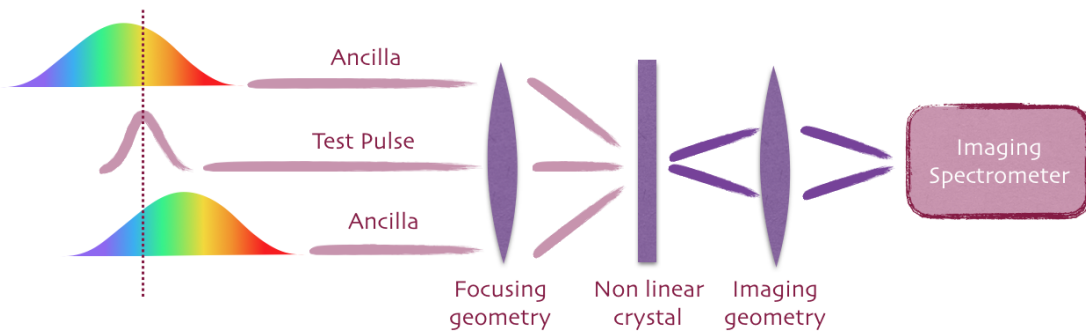


Fig. 3.13 SEA-SPIDER scheme. The test pulse is upconverted with two monochromatic slices of two time-delayed temporally-chirped ancillae. The geometry used is non collinear, hence the signal will display spatial fringes, which are resolved with an imaging spectrometer.

A scheme of the setup is described in Fig. 3.13. In this technique two spectrally sheared replicas are created by upconverting the TP with two time-delayed not collinear chirped ancillary pulse. Using two ancillas allows for less manipulation of

the test pulse (TP) compared to the general SPIDER. The two TP replicas are not delayed in time, and thus there are not any spectral fringes: in conventional SPIDER in order to correctly resolve the interference fringes in the frequency domain, the spectral sampling rate needs to be higher than that imposed by the WS sampling theorem, which results in added noise in the reconstruction. In SEA SPIDER instead the phase information is encoded in spatial fringes, allowing the spectral sampling rate to be at the sampling limit imposed by the WS theorem. The signal detected by an imaging spectrometer is given by:

$$S(x, \omega) = |E(x, \omega - \omega_0)|^2 + |E(x, \omega - \omega_0 - \Omega)|^2 + \\ + 2|E(x, \omega - \omega_0)||E(x, \omega - \omega_0 - \Omega)| \cos[\phi(x, \omega - \omega_0) - \phi(x, \omega - \omega_0 - \Omega) + Kx]$$

where K is the difference between the transverse component of the propagation vectors. A typical interferogram is shown in the inset of Fig. 3.13. The phase reconstruction process is the same as in conventional SPIDER, but a 2D Fourier transform (and 2D inverse Fourier transform) is performed to isolate the sideband peak. The resulting function is :

$$S^{AC}(x, \omega) = |E(x, \omega - \omega_0)||E(x, \omega - \omega_0 - \Omega)| e^{[i\phi(x, \omega - \omega_0) - i\phi(x, \omega - \omega_0 - \Omega)]} e^{(iKx)}$$

the calibration phase Kx can be removed by performing a measurement without shear. Processing independently each spatial slice of the interferogram with the same SPIDER algorithm leads to the reconstruction as for each slice the spatial phase is constant and thus it only adds to the unretrieved value of the absolute

phase. Since this is the case however no details can be obtained on the space-time coupling of the TP as this technique provides no wavefront information.

3.2.1.3. SEA-CAR-SPIDER

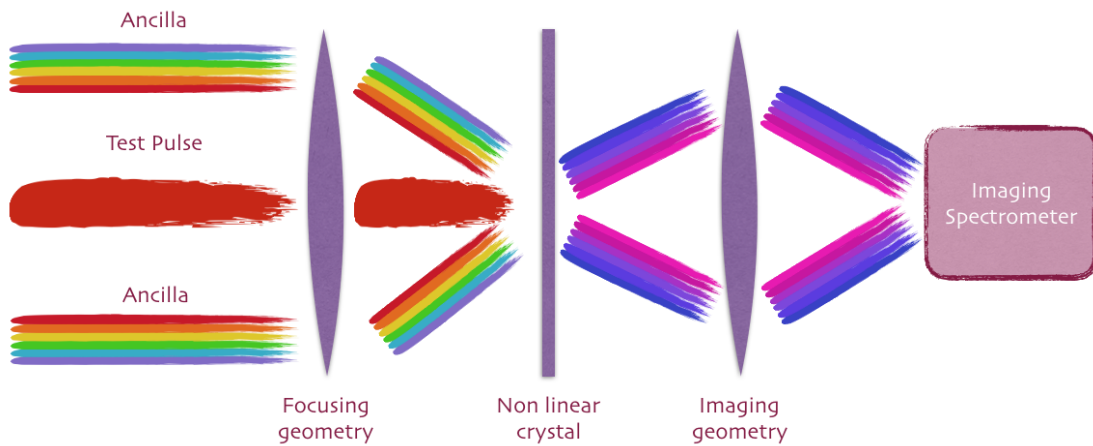


Fig. 3.14 SEA-CAR-SPIDER scheme. The test pulse is upconverted with two spatially chirped ancillae (with opposite chirp). The signal is then recorded with an imaging spectrometer.

One of the major limits of conventional SPIDER is the impossibility to reconstruct spectra which exhibit spectral nulls wider than the shear. This can be solved by using larger and larger shears, but as the resolution become worse, the temporal window allowed by the WS theorem is reduced as well. It is possible to acquire data for conventional SPIDER reconstruction at different shears with multiple runs, but that would require choosing the shear in advance and is not suitable, as all non-single shot techniques, if the source is not particularly stable. A solution to this problem is offered by SEA-CAR SPIDER [21, 23], in which multiple shear reconstruction is performed in a single shot measurement. This is possible by spatially chirping the ancillary pulses, with opposite chirps. The unknown pulse is upconverted in each position with a different monochromatic slice of each ancilla, and

the test pulses will then have opposite spatial chirp as well, with a linearly varying frequency in space. The two test pulses are imaged into an imaging spectrometer forming a relative angle θ which allows interference in the spatial domain. The shear will be given by $\Omega = \omega_A - \omega_B = 2\alpha x$ where alpha is the amount of chirp, and thus it changes with the position. The setup scheme is reported in Fig. 3.14. The resulting signal reads:

$$S(x, \omega) = |E(\omega - \omega_A)|^2 + |E(\omega - \omega_B)|^2 + \\ + 2|E(\omega - \omega_A)||E(\omega - \omega_B)| \cos \left[\phi(\omega - \omega_B) - \phi(\omega - \omega_A) + \frac{\theta \omega_{up} x}{c} \right].$$

As in the SEA SPIDER case the phase can be reconstructed for each spatial position, and consequently for different amounts of shear. This redundancy can be used in specific multishear algorithms to improve the precision of the reconstruction [22]. Another advantage of this technique is that there is no need to take additional measurements to remove the calibration phase as the trace already includes a position in which the shear is zero, and reconstructing the phase in that position would give the reference phase.

3.3. THE MICE ALGORITHM

The use of a self-referenced technique opposed to a referenced one is usually preferred in ultrafast metrology as a stand-alone method is more flexible, not needing a reference to be "adjusted" to the pulses under study. Furthermore the strong drawback of having to know the reference a priori and to use it in order to characterize the pulses desired, can also lead to a strong propagation of the reconstruction error as it adds one step to the reconstruction which introduces further inaccuracies. However, the use of a reference has also many advantages

which can be a key point in many characterisations: for example it can provide more power when the pulses to be analysed are particularly weak, it can increase the sensitivity and provide an overall better SNR, and can be particularly helpful in those cases where frequency doubling is not an option and other types of mixing can be preferred.

Together with Dr. C. Bourassin-Bouchet and Dr. M. M. Mang, I have developed a new technique which allows for the benefits of a referenced techniques avoiding anyway the drawbacks just stated. This new technique, which we christened Mutual Interferometric Characterisation of Electric-fields (MICE) has been published in Optics Letter [96] and achieved a special mention in Spotlight on Optics of December 2014.

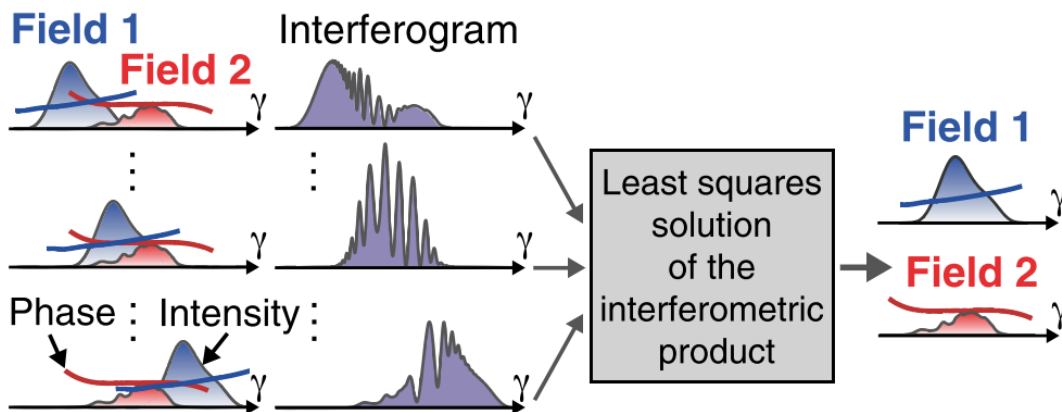


Fig. 3.15 MICE scheme. Two complex fields, depending on a parameter γ are sheared multiple times and the interferograms are recorded. The MICE algorithm retrieval based on a least squares iteration allows for the simultaneous retrieval of both fields.

MICE exploits the capabilities of the multishear approach which guarantees the redundancy of information needed for reconstructing more than one field at once. The more shears are used, the more fields can be reconstructed, posing virtually no limits to the number of simultaneous reconstructions. Using this technique it is thus

possible to have a beam which acts as a reference without being known a priori, supplying all those features which might be lacking in the beam under study, as for example power or bandwidth. In order to explain the mechanism behind MICE we consider the case of having only two fields, $E_1(\omega)$ and $E_2(\omega - \Omega)$, where E_2 has been sheared of the amount Ω . The interferogram resulting from the interference of these two fields can be written as:

$$I_{jk} = |E_1(\omega_j)|^2 + |E_2(\omega_j - \Omega_k)|^2 + 2\text{Re}(E_1(\omega_j) E_2^*(\omega_j - \Omega_k)) \quad (3.14)$$

As in SPIDER the interference term $E_1(\omega_j) E_2^*(\omega_j - \Omega_k)$ is isolated via Fourier transforming the interferogram and selecting one AC band, namely $AC_{j,j-k}^{meas}$. In order to separate the contribution from the two different fields, a least square approach is used. Defining the error:

$$\epsilon = \sum_{j,k}^{J,K} |AC_{j,j-k}^{meas} - E_1(\omega_j) E_2^*(\omega_j - \Omega_k)|^2, \quad (3.15)$$

it is possible to obtain the two fields by minimizing it with respect to both E_1 and E_2 in an iterative algorithm. In order to minimize the error, one must set to zero the derivatives:

$$\begin{aligned} \frac{\partial \epsilon}{\partial E_1(\omega_j)} &= 0 \\ \frac{\partial \epsilon}{\partial E_2(\omega_j - \Omega_k)} &= 0 \end{aligned} \quad (3.16)$$

which leads to the following equations:

$$E_1(\omega_j) = \frac{\sum_k AC_{j,j-k}^{meas} \cdot E_2(\omega_j - \Omega_k)}{\sum_k |E_2(\omega_j - \Omega_k)|^2}$$

$$E_2^*(\omega_j) = \frac{\sum_k AC_{j,j+k}^{meas} \cdot E_1^*(\omega_j + \Omega_k)}{\sum_k |E_1(\omega_j + \Omega_k)|^2}. \quad (3.17)$$

These equations are solved in an iterative algorithm starting with a random guess for one of the fields which is used to obtain a solution for the other field which is then used to improve the solution for the other field, until the error is minimized. In this way each field acts as a reference for the other, hence this can be considered a mutually-referenced technique.

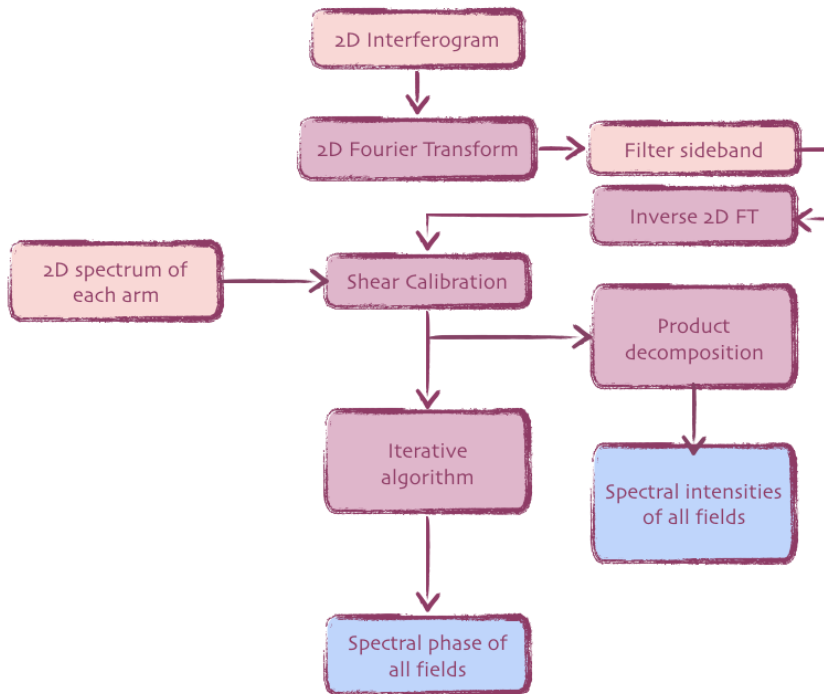


Fig. 3.16 MICE block diagram

The convergence is usually very fast requiring only a few steps to reach a very good agreement between the retrieved AC band and the measured one. This is due to the fact that contrary to the FROG iterative algorithm, in MICE, the phase information is encoded in the fringe pattern as in all interferometric methods. This intrinsic difference also reduces the number of ambiguities to which MICE is subject compared to methods like blind-FROG. In fact in the case where only two fields are present, MICE suffers only from the following ambiguity:

$$E_1(\omega_j) \cdot E_2^*(\omega_j - \Omega_k) = \alpha E_1(\omega_j) \cdot \frac{1}{\alpha} E_2^*(\omega_j - \Omega_k), \quad (3.18)$$

where α is a complex number. This results in an indetermination of the absolute intensity and phase. While the latter is common in all the known reconstruction methods, the intensity ambiguity can be resolved by measuring individually the spectra of the two fields or by using the DC component of the interferogram where the symmetry between the two fields is broken.

In order for this algorithm to lead to a successful reconstruction there needs to be redundant information in the interferogram, i.e. the greater the number of shears considered, the lesser noise will be present in the reconstruction. Furthermore the fields will be reconstructed only in the region covered by the shears, hence the shears need to be chosen so that two subsequent interfering regions of the interferogram overlap. As stated earlier an arbitrary number of fields can be reconstructed at the same time, but that would consequently require a greater number of shears to be used leading to an experimental limit ultimately due to the instrument's resolution. Furthermore more ambiguities will be present when more fields are considered, as it happens when adding a third field in the equations, which we will see in the following paragraph. This technique proved to be very versatile and can be used for an extensive range of applications. The MICE technique can in fact be adapted to

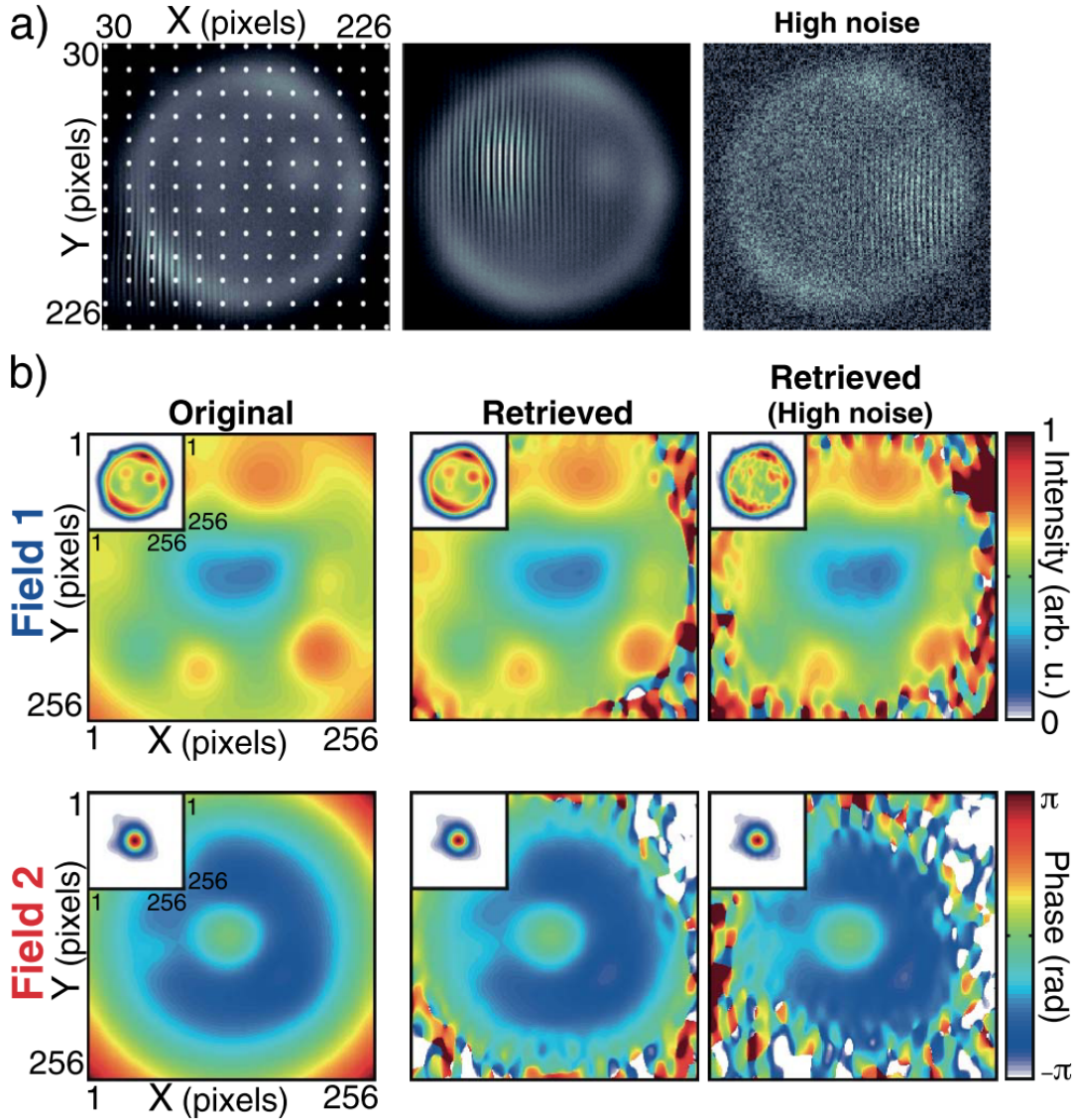


Fig. 3.17 (a) Spatial wave front reconstruction. Interferograms for three different shears. The white dots represent the locations of the different spatial shears used for the reconstruction. The interferogram in the right panel represents the high noise case. (b) Simulated and reconstructed wavefronts of both fields. Insets: Spatial intensity profiles of the two fields.

many scenarios not only linked to the temporal reconstruction, e.g. it can be used also to reconstruct the spatial profile of a beam utilizing a spatial displacement instead of the frequency shear Ω . An example of this can be found in Fig. 3.17 of a simulated 2D Waveform reconstruction.

3.3.1. APPLICATION TO SEA-CAR SPIDER

MICE can be used with any multishear technique respecting the criteria mentioned in the previous paragraph, and thus SEA-CAR SPIDER is an optimum candidate to test its potential. However as seen before SEA-CAR SPIDER uses two ancillary beams to apply a spatial shear to the test pulses, which can be described as two complex functions varying only with the shear $a_{1,2}(\Omega_{1,2})$ hence needing to adjust the MICE equation accordingly. Given the electric fields $E_{1,2}(\omega' - \Omega_{1,2})$ and the ancillary fields just discussed, using the variable change $\omega = \omega' - \Omega_1$, it is possible to write the AC product as $E_1(\omega_j) E_2^*(\omega_j - \Omega_k) A(\Omega_k)$, where $A(\Omega_k) = a_1 a_2^*(\Omega_1 - \Omega_2)$ resulting in the equivalent of a three-field scenario.

The equations to solve for minimising the error then become:

$$\begin{aligned}
 E_1(\omega_j) &= \frac{\sum_k AC_{j,j-k}^{meas} \cdot E_2(\omega_j - \Omega_k) \cdot A^*(\Omega_k)}{\sum_k |E_2(\omega_j - \Omega_k) \cdot A(\Omega_k)|^2} \\
 E_2^*(\omega_j) &= \frac{\sum_k AC_{j,j+k}^{meas} \cdot E_1^*(\omega_j + \Omega_k) \cdot A^*(\Omega_k)}{\sum_k |E_1(\omega_j + \Omega_k) \cdot A(\Omega_k)|^2} \\
 A(\Omega_k) &= \frac{\sum_j AC_{j,j-k}^{meas} \cdot E_1^*(\omega_j) \cdot E_2(\omega_j - \Omega_k)}{\sum_j |E_1(\omega_j) \cdot E_2(\omega_j - \Omega_k)|^2}.
 \end{aligned} \tag{3.19}$$

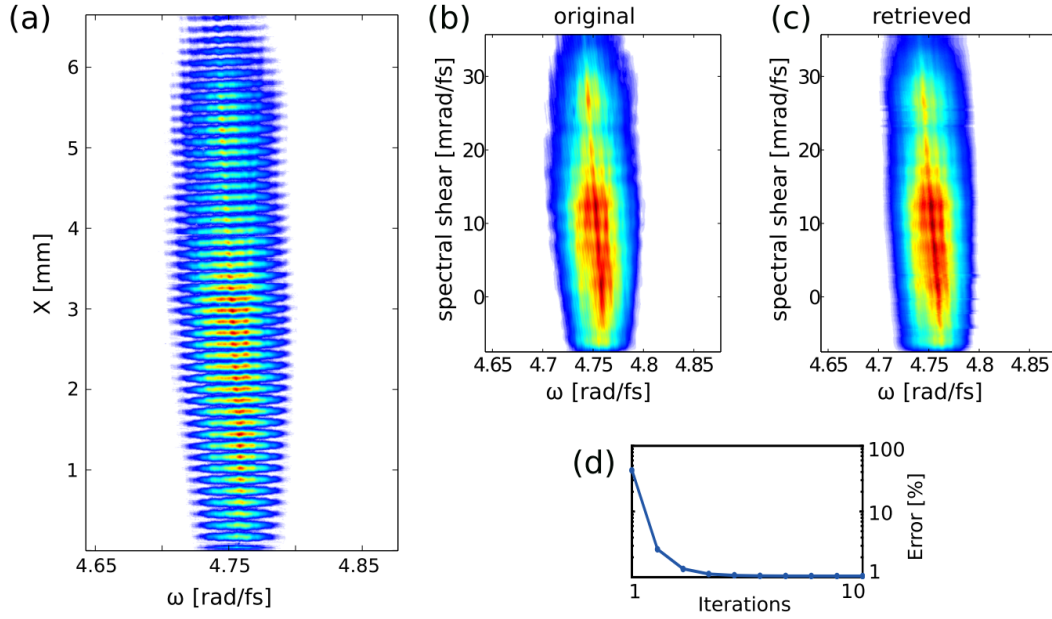


Fig. 3.18 (a) MICE interferogram of two pulses; moduli of the original (b) and retrieved (c) interferometric products associated to the interferogram in (a) after 10 iterations of (3.19). (d) Evolution of the error between the original and retrieved interferometric products.

As for the two-fields case, these equations can be solved iteratively starting with a random guess for two of the three fields involved instead of one. However, the addition of one field results in a second ambiguity:

$$E_1(\omega_j) \cdot E_2^*(\omega_j - \Omega_k) \cdot A(\Omega_k) = E_1(\omega_j) e^{\beta\omega_j} \cdot E_2^*(\omega_j - \Omega_k) e^{-\beta(\omega_j - \Omega_k)} \cdot A(\Omega_k) e^{-\beta\Omega_k}, \quad (3.20)$$

where β is a complex number. The real part of β , as α , contributes to the intensity ambiguity and can be resolved by a direct measurement of the spectra; the imaginary part of β leads to an ambiguity in the phase linear term, however the fields are defined up to an arbitrary delay, so this ambiguity is irrelevant and it does not affect the information on their relative delays, i.e. preserving their relative phases.

An example of such reconstruction is shown in Fig. 3.18 where the reconstruction error is shown together with the interferogram and the retrieved and measured AC band. From the dependence of the reconstruction error on iterative step number it is possible to appreciate the rapid convergence of the algorithm with the error going below 2% after only three steps of the algorithm.

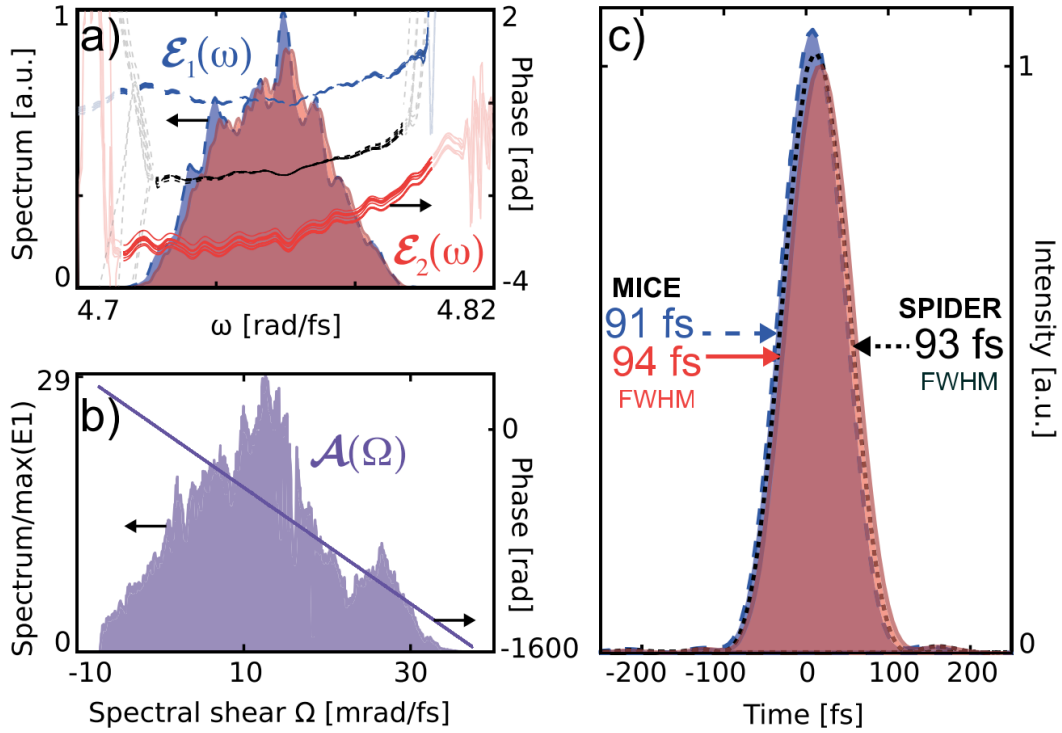


Fig. 3.19 (a) Fields E_1 (dashed blue line), E_2 (red line), and (b) A retrieved for 10 single-shot measurements. (c) Single-shot pulse profiles of the fields E_1 (dashed blue line) and E_2 (red line). The black dotted lines stand for the spectral phase (a) and pulse profile (c) retrieved with SPIDER.

The three reconstructed fields are shown in Fig. 3.19 where it is possible to observe the difference between the pulses obtained with MICE and the SPIDER typical reconstruction. As expected the two pulses look almost identical however it is possible to notice a slight difference in peak intensity and a delay between the two pulses of $\simeq 10$ fs.

3.4. MIND THE GAP

The core objective of this thesis is to find a metrology technique that can be successfully employed for the reconstruction of ultrashort pulses with a spectrum consisting of two or more separate spectral components, i.e. a spectrum which exhibits one or more spectral gaps. In particular we define a *spectral gap* as a lack in spectral intensity on a support $\Delta\omega_g$ which is larger than the support of each spectral component $\Delta\omega$.

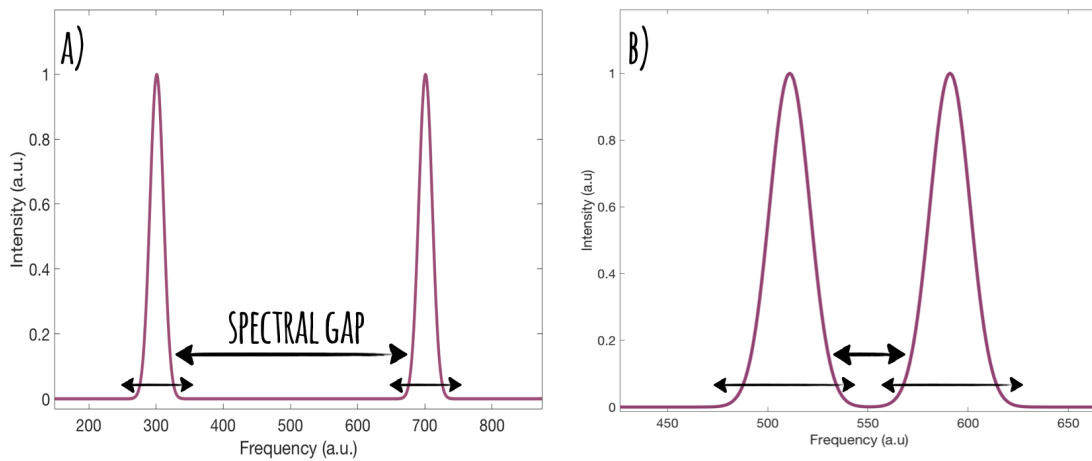


Fig. 3.20 An example of a spectral gap a) larger than the components support, b) smaller than the components support

If in fact this is not the case and the gap is smaller than one of the components as in Fig. 3.20 b), there are many established techniques which allow us to characterize such pulses, for example the SEA-CAR-SPIDER approach can be successful with the right choice of shears [23], as we will shortly discuss. Measuring pulses with a spectral gap so defined instead is a long standing problem which has been the subject of studies for the past decade [23, 97, 98]. In particular in [97] Keusters et al. provide a thorough discussion on how common self referenced techniques

fail to reconstruct pulses with well-separated components due to a relative phase ambiguity. While it can be fairly easy to measure all the individual components on their own, finding a technique which is capable of retrieving the relative phase of well separated components without additional requirements or constraints on the pulses is non trivial and is the objective this thesis work aims to achieve.

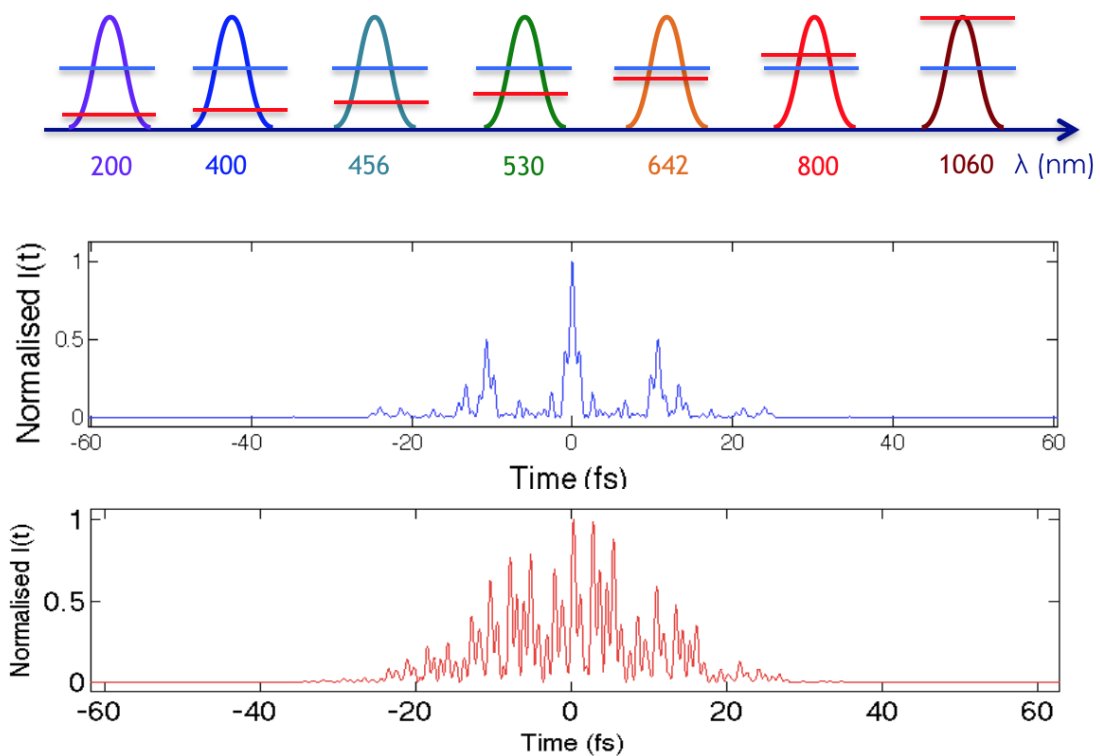


Fig. 3.21 Temporal profile of a pulse with varying relative phase. In both cases the phase of each individual component is flat, but while in the first case (blue) the relative phase is set to zero, in the second one (red) the relative phase increases quadratically.

The relative phase between the spectral components is crucial in determining the time duration of the pulse which is strongly affected by relative phase variations as shown in Fig. 3.21: in the example given, the temporal profile of a pulse with a spectrum composed of many well-separated spectral components each with a flat spectral phase is calculated in the instance in which the relative phase between all

of them is set to zero (blue), or it exhibits a quadratic behaviour (red). While the blue case corresponds to the Fourier limited scenario, and the temporal FWHM of the central peak is 700 as, when the relative phase is set to increase quadratically the pulses duration can change dramatically (to a few tens of fs, in the example given). The difference in the time duration is solely due to the relative phase change is such that it is evident that simply knowing the phase of the individual spectral components gives only partial information on the pulse. Thus, it is essential to establish a method that is able to provide reliable information about the relative phase between spectrally separate components of the field.

We discuss in the following sections why common methods fail in retrieving this relative phase underlining the ambiguities which could result from any characterisation attempt with those techniques. Determining the relative phase can be important in a range of different applications [99, 100] and even more prominently in all the light sources which are intrinsically characterised by a gap structure are discussed later in this chapter.

3.4.1. SPIDER

As we have seen earlier SPIDER – and especially its SEA-CAR variant, could be considered good candidates for treating spectral gaps as they successfully retrieves pulses with smaller gaps. However what limits these techniques is a property of the reconstruction algorithm itself and thus it cannot simply be extended to the case of arbitrarily larger gaps.

Specifically, what fails the reconstruction of gapped pulses is the concatenation step: the phase, sampled with the shear, is constructed by summing up on two consecutive sample points; if one point happens to be on a zero-intensity spot (i.e. a spectral gap), all the previous information on the phase will be lost and the

successive reconstruction will continue starting from a random value the phase will get in that point, until it meets again a zero-intensity spot, which then resets to a random value again. Thus, the concatenation fails to reconstruct the phase with continuity in all instances where a sampling point falls in a zero-intensity spot. Hence, if using a self-referenced, single-shear version of SPIDER, this prevents the complete reconstruction with any gap larger than the sampling rate: it is still possible to use SPIDER to successfully reconstruct the phase of the each spectral component, but their relative phase would remain unknown. However this can be slightly relaxed if using a multishear approach or a cross-referenced technique:

- **Multi-shear.** The multi-shear approach can successfully reconstruct gapped pulses provided that the gap is smaller than the support of the components. If that is the case in fact, as shown in Fig. 3.22, for a particular shear it is possible to get the interference between both peaks (in the case of a pulse with two spectral features, but can be easily extended to a more general case). At the same time a smaller shear is used to reconstruct the phase along each spectral component. This has been done by Austin *et al.* in [23].
- **Cross-Referenced technique.** A different approach involves using a known reference pulse, as it is done with the XSPIDER technique [94, 95]. The reference needs to have a bandwidth larger than that of the gapped pulse, and must be characterized in advance. Also in this case there is need to use a multishear approach: a larger shear to span the gap and a smaller one to retrieve the individual components phases.

If the gap is larger than the support of the spectral features it is impossible to find a shear to span the gap, as even with the largest possible shear one in two consecutive sampling points would fall within the gap. As it will be discussed in

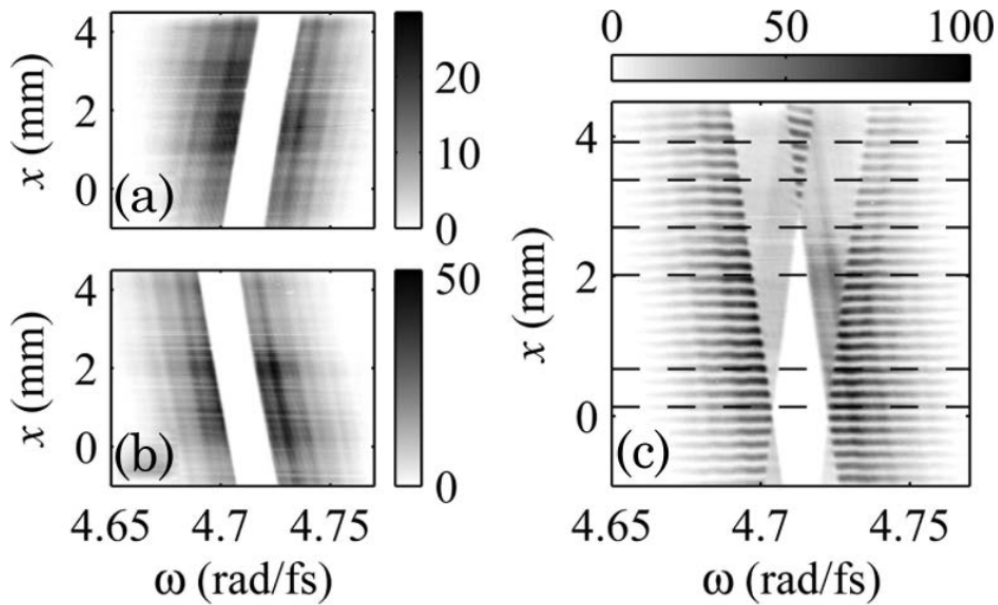


Fig. 3.22 SEA-CAR-SPIDER gap interferogram [23]. (a) and (b): Individual spectra of the upconverted replicas; (c): SEA-CAR-SPIDER interferogram.

5.2.1 a further limitation in SEA-CAR-SPIDER is due to the upconversion, where an additional phase needs to be taken into account and prevents the method from reconstructing well-separated components. On the other hand, the cross referenced approach is not easily applicable to cases with many gaps, especially if the gaps have different widths as that would require different shear values, and thus measurement, for each gap. Furthermore the reference to be used would need to be very well characterized to avoid increasing errors in the reconstruction. This latter point can be complicated if the bandwidth required is particularly large. However, both the techniques considered serve to underline how a multishear approach can be used for characterising complex pulses if one can overcome the multiple challenges and intrinsic limitations described above.

3.4.2. FROG

Self-referenced FROG methods, as for example SHG-FROG fail in the gap reconstruction because, similarly to SPIDER, the information between the spectral components is lost. The simulation in Fig. 3.23 shows how the SHG-FROG trace of a gapped pulse results in exactly the same trace for two different values of the relative phase between the spectral components.

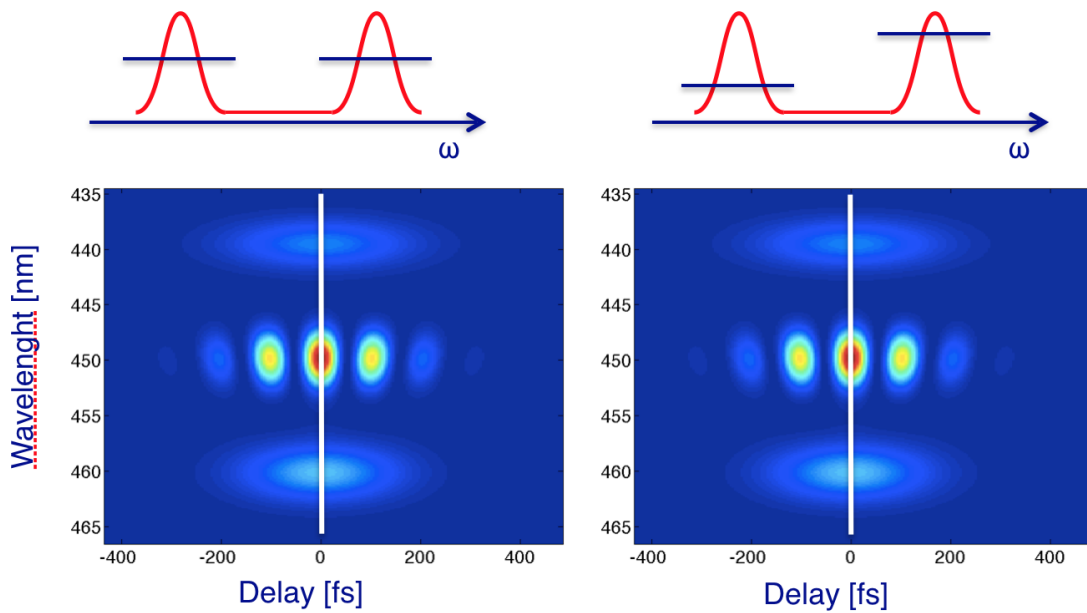


Fig. 3.23 Simulation of a SHG-FROG for a pulse with a spectral gap with zero relative phase (left) and a π shift between the two spectral components (right). The SHG-FROG traces in the two scenarios are identical, hence the information on the relative phase is not conveyed by the trace.

As for the interferometric equivalent, the natural step forward is to consider the cross-referenced version of FROG (XFROG) [16]. However for this technique to work the reference must be correctly shaped both in the frequency and in the time domain, and needs to have the right amount of chirp to match the incoming gapped pulse. Such a tailored reference would then need to be characterized correctly for each new pulse to be tested. As shown in the simulation of Fig. 3.24, when the

reference used covers the gap bandwidth it is possible to discriminate between the two spectrograms with different phases.

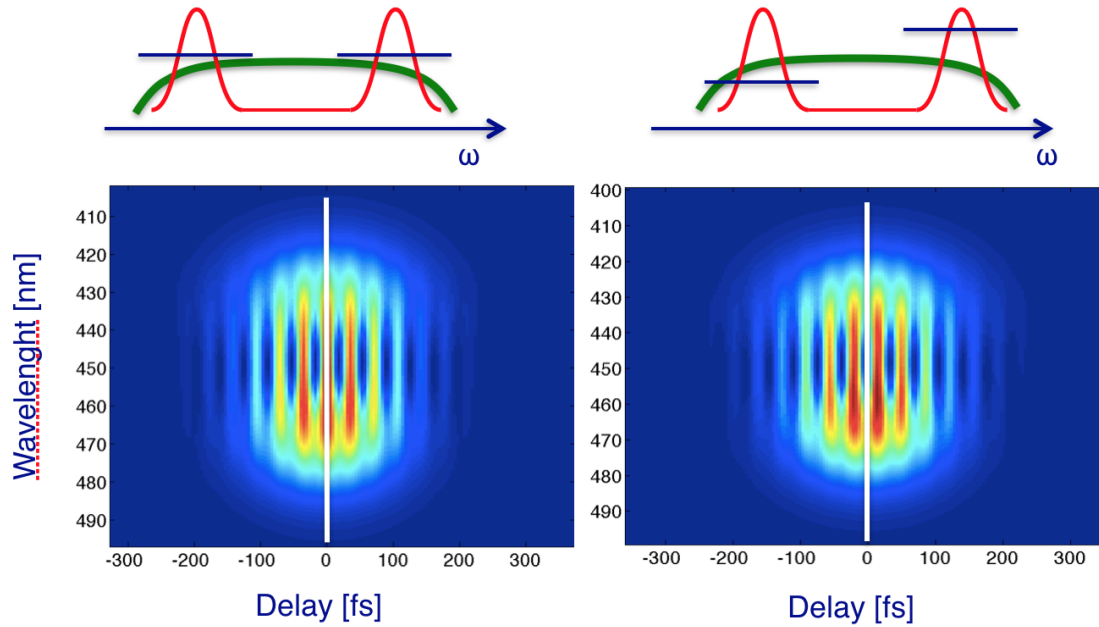


Fig. 3.24 Simulation of an X-FROG trace between a pulse with a spectral gap and one with a bandwidth larger than the gap with zero relative phase (left) and a π shift between the two spectral components (right). In this case the FROG traces look different and it is possible to discriminate between the two scenarios.

Despite the difficulties of having a broad, tailored reference, it is however possible to use XFROG to characterise the single spectral components, and some preliminary information about the pulse can be extracted in this way as it is shown in Chapter 4.

A different approach would be to use the blind-FROG technique [75], which utilizes the PCGPA algorithm to retrieve both the probe and gate pulses and thus does not require the use of a known reference for the characterisation. This technique however is subject to many nontrivial ambiguities, as for example described by Seifert et al. in [101], where it's shown that blind-FROG leads to a unique solution only if the spectral intensities of the two pulses are known and the trace is non-centrosymmetric. This adds up to the relative-phase ambiguity discussed in [97],

which is common to all techniques which do not use an external well-characterized reference. To solve these ambiguities Seifert et al. proposed a modified blind-FROG variant, called Very Advanced Method for Phase and Intensity Retrieval of Electric-fields (VAMPIRE) [98]. In VAMPIRE the input pulses are manipulated to always obtain a non-centrosymmetric trace: this is achieved by applying a spectral phase modulator on one of the two input signals to avoid the centrosimmetry of the trace and by assuring that the same signal is broad enough to cover the spectrum of the gapped pulse and does not itself have gaps. This can be either obtained by using an independent pulse or by using a temporal phase modulator as well as a spectral phase one. This method has been used to successfully characterise soliton molecules [102]. However for VAMPIRE to perform correctly it is required that the spectra are known and the conditioning filter applied is fully characterized.

CHAPTER 4

CHARACTERIZATION OF COMPOSITE OCTAVE-SPANNING FREQUENCY COMBS

Whaddya think it is, MICE?

Well, dummy, think twice!

[...] Frogs! We're the frogs!

The adorable frogs!

The Frogs, Stephen Sondheim

4.1. INTRODUCTION

In this chapter I describe our first approach to the characterization of spectral-gap pulses. This has been the result of a collaboration with Prof. D. Reid's group in Heriot-Watt University. Their new approach to frequency comb generation called for a way to measure the duration and most importantly the relative phase between the discrete spectral components of their pulses. In what follows I will give a brief background to describe the type of pulses developed and focus on the features of

the XFROG setup, that I designed and developed, and how we used it to obtain insightful information for improving pulses compression and overlap. The results obtained have been published on Optics Letters [103].

4.2. COHERENT SYNTHESIS

Coherent pulse synthesis consists in crafting arbitrarily shaped optical pulses by combining mutually coherent sources. The sources used can vary, for example Ti:Saph lasers [27] and optical parametric amplifiers [53] have been used to obtain respectively ultra-broadband and sub cycle pulses. At Heriot Watt University a new source has been developed, based on the use of a Carrier envelope phase stabilized Optical parametric oscillator (OPO) [54]. An OPO delivers a frequency comb given by the possible combinations between the pump, signal and idler pulses [104–106].

In order to successfully obtain coherent pulse synthesis two conditions need to be met:

- 1 the parent pulses need the same repetition frequency f_{rep} ;
- 2 the parent pulses need to have a common CEO frequency;

The OPO automatically satisfies the first requirement as all the components come from the same and only source and thus share the same f_{rep} . The second condition instead needs to be imposed and the only possible solution is that all the CEO frequencies are locked to zero, in fact, any nonlinear combination between the pump signal and idler, given that $f_{CEO}^p = f_{CEO}^s + f_{CEO}^i$, can be expressed by:

$$f_{NL} = \alpha f_{rep} + \beta f_{CEO}^p + \gamma f_{CEO}^s, \quad (4.1)$$

where α, β, γ are integers f_{rep} is the repetition frequency and $f_{CEO}^{p,s}$ are the CEO frequency of the pump and signal respectively. Thus, if the CEO needs to be common for each parent pulse implies

$$\beta f_{CEO}^p + \gamma f_{CEO}^s = \beta' f_{CEO}^p + \gamma' f_{CEO}^s = \beta'' f_{CEO}^p + \gamma'' f_{CEO}^s \text{ etc,} \quad (4.2)$$

which is satisfied only if $f_{CEO}^p = f_{CEO}^s = f_{CEO}^i = 0$ in the general case.

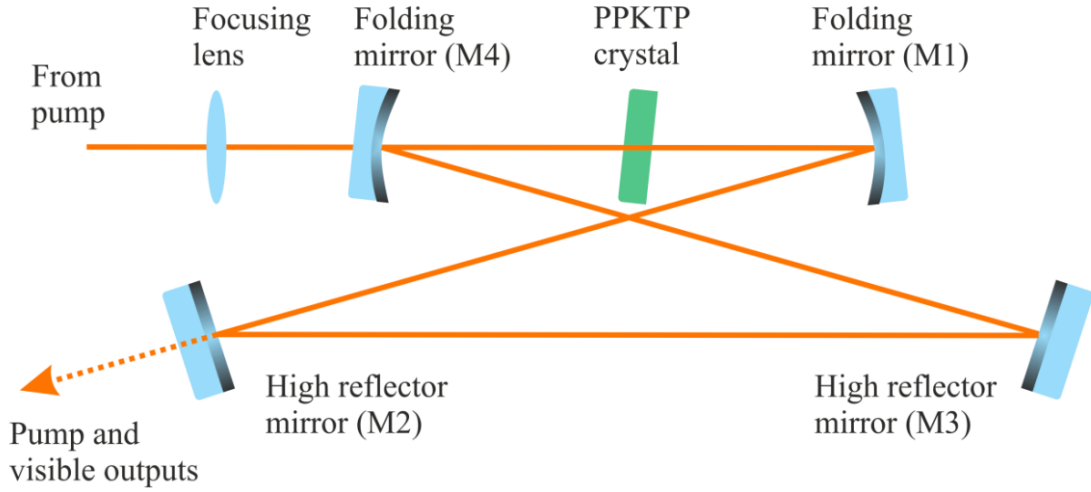


Fig. 4.1 Experimental setup for the generation of a composite frequency comb

The experimental setup for the coherent source is described in Fig.4.1. The pump pulse coming from a Ti:Saph enters in a ring cavity in which a PPKTP crystal is placed, which acts as the OPO.

The PPKTP crystal is mounted on a "Pac-man" style mount to allow maximum clearance for beam steering and put on a 3D translation stage. The crystal was tilted so that the front and back reflection of the pump could be blocked with a pinhole. The OPO cavity was initially designed with a linear geometry but that led to very low powers reported in Table 4.1. The spectrum obtained is composed of

Pulse	Wavelength (nm)	Linear Cavity Powers (mW)	Ring Cavity Powers (mW)
2p	400	3	10
ps	450	8	30
2s	530	45	100
pi	630	not measurable	4

Table 4.1 Powers comparison between linear and ring cavity arrangement

the pump (p) pump-signal mixing (ps), pump-idler mixing (pi). double pump (2p) and signal doubling (2s).

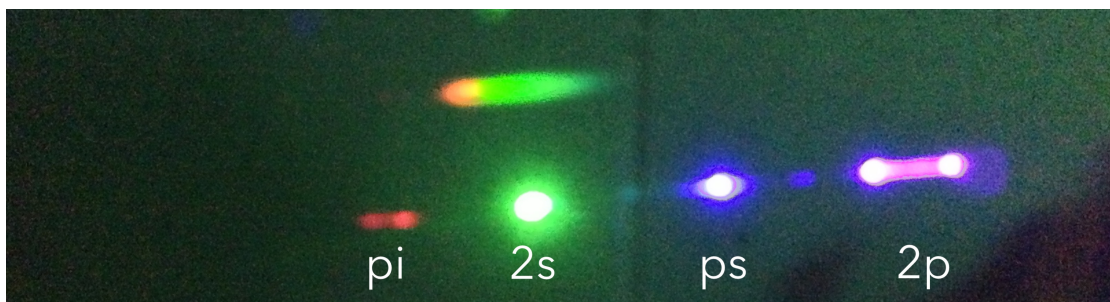


Fig. 4.2 Spectrum of the OPO diffracted off a prism. It is possible to clearly distinguish the main four different frequency mixing from the OPO at 630 nm, 530 nm, 450 nm and 400 nm.

4.3. COMB CHARACTERIZATION WITH XFROG

The source just described poses three main challenges for its characterization:

1. **Gap structure:** its spectrum is fragmented and shows gaps wider than the spectral components.
2. **Large bandwidth:** the bandwidth covered ranges through all the visible spectrum.
3. **Low powers:** as seen in Table 4.1, the powers of the different colours can be quite low for some of the frequency-mixing signals.

In Chapter 3 the limitations that current techniques face when dealing with gap structure are discussed. Nonetheless some of these techniques can be useful to obtain some preliminary information about the pulses, despite failing in allowing a full reconstruction. One technique capable to do so is XFROG. The initial idea was to perform an XFROG measurement between a small self phase modulated (SPM) portion of the pump beam and all the colours. If the SPM was broad enough to span even the largest gap, this would allow to retrieval of the relative phase and obtain a full reconstruction of the pulse, provided that the SPM was broad enough also in the time domain and that it could be fully characterized before. However this proved to be impossible due to the low powers of the pulses and it appeared clear that we needed to fully utilize the XFROG capabilities to get as much power as possible from the pump pulse to compensate for the low power of the different components. This is also due to the choice of the nonlinear crystal: if the bandwidth was smaller one could have chosen a thicker crystal to compensate for the lack of power, but with such a bandwidth only a $10\ \mu\text{m}$ long crystal could be adopted to cover the whole OPO range as in Fig 4.3, leaving a very short interaction length to be utilized. This needed of a trade off: in order to get more signal we opted for a $30\ \mu\text{m}$ instead, which allowed a few components of the OPO to be observed but with enough power for detection.

Instead of using SPM we proceeded to use XFROG for all the colours with the pump: this is sufficient to correct for the GD between the components and their compression but does not give any info about the relative phase, which in the reconstruction analysis that follows is assumed to be zero.

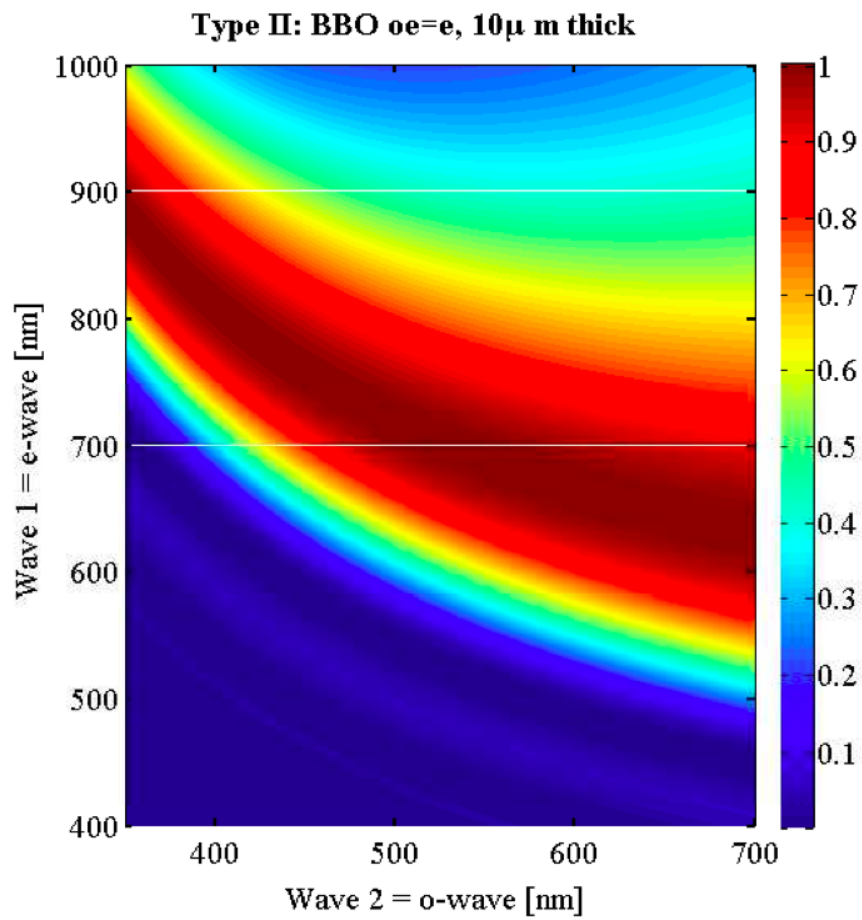


Fig. 4.3 Phase-matching function, normalized to its maximum, with a 10 μ m Type II BBO crystal

4.3.1. DESIGN

The setup for XFROG is shown in Fig. 4.4. This setup has been carefully designed to deal with the low powers provided by the visible pulses of the OPO.

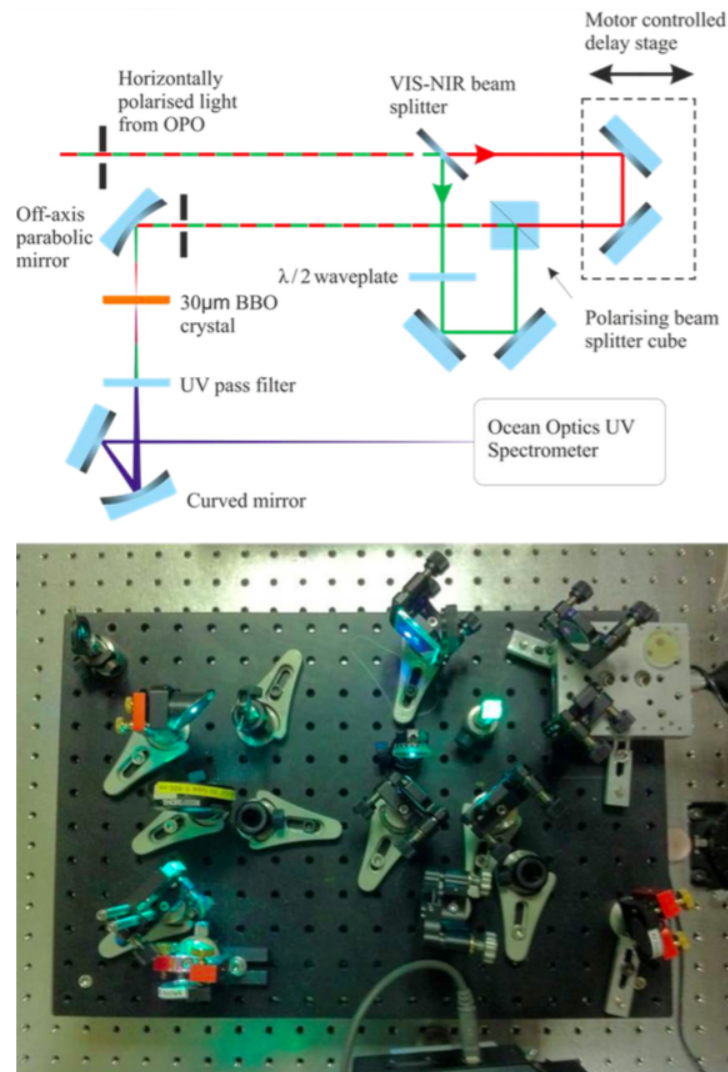


Fig. 4.4 Experimental scheme for the XFROG setup. The light coming from the OPO is split between the pump (red) and visible pulses (green) via a visible-NIR beam splitter and then recombined using a polarizing beam splitter (PBS). The beam is then focused with a parabolic mirror onto a Type II 30 μm BBO crystal cut at 45.6° . The signal is then recorded by means of an Ocean Optics UV Spectrometer

The pump beam and the visible pulses enter collinearly in the setup and are split by means of a visible – NIR beam splitter into the two arms. Obviously there are some losses in the process, meaning that part of the pump will be reflected together with the colours of the OPO allowing one to also measure the pump itself. The pump, which is horizontally polarised, is then used as a gate. The mirrors on the pump arm are mounted on a closed loop piezo stage which allows scanning the delay between the two arms. The visible pulses' polarization, previously horizontal too, is rotated by means of a half wave plate (HWP) at 45° so to become vertically polarized and the visible pulses are then recombined with the pump using a polarization beam splitter cube (PBS). The signal is focused in the crystal by means of a 90° parabolic off-axis mirror with a focal length of 50 mm. The use of the parabolic mirror instead of a spherical mirror is due to the fact that having such a big reflection angle allow us to place a camera in the focus to check the spatial alignment not only in the near but also in the far field. This is not relevant when the powers of the two arms are high but with low power the spatial overlap condition that is necessary for efficient upconversion process becomes critical. For getting the second harmonic generation signal we use a $30\ \mu\text{m}$ long BBO crystal cut at 45.6° for Type II (oe $\rightarrow$ e) SFG between the pump and the visible pulses. The resulting signal was then filtered with a UV-pass filter and imaged into an Ocean Optic spectrometer that covered the spectral range of 227–450 nm.

4.3.2. ALIGNMENT

Careful alignment of the two arms was very critical in assuring that the XFROG setup would work with the low power exhibited by the OPO visible pulses. The crucial step resulted to be that the pump and visible pulses were collinear and perfectly overlapped when focusing through the SFG crystal to maximise the SFG

signal. In order to achieve such overlap a webcam was used in the focal plane of the off-axis parabolic mirror and in the far field. To reduce the power on the webcam a variable ND filter was placed in the beam path before the XFROG setup. In Fig. 4.5 a typical image from the webcam in the focal plane is shown. The pump beam, in which the steering mirrors were fixed on the piezo stage, was used as a reference. The visible beam instead could be steered until the focal spots overlapped (left side of Fig 4.5).

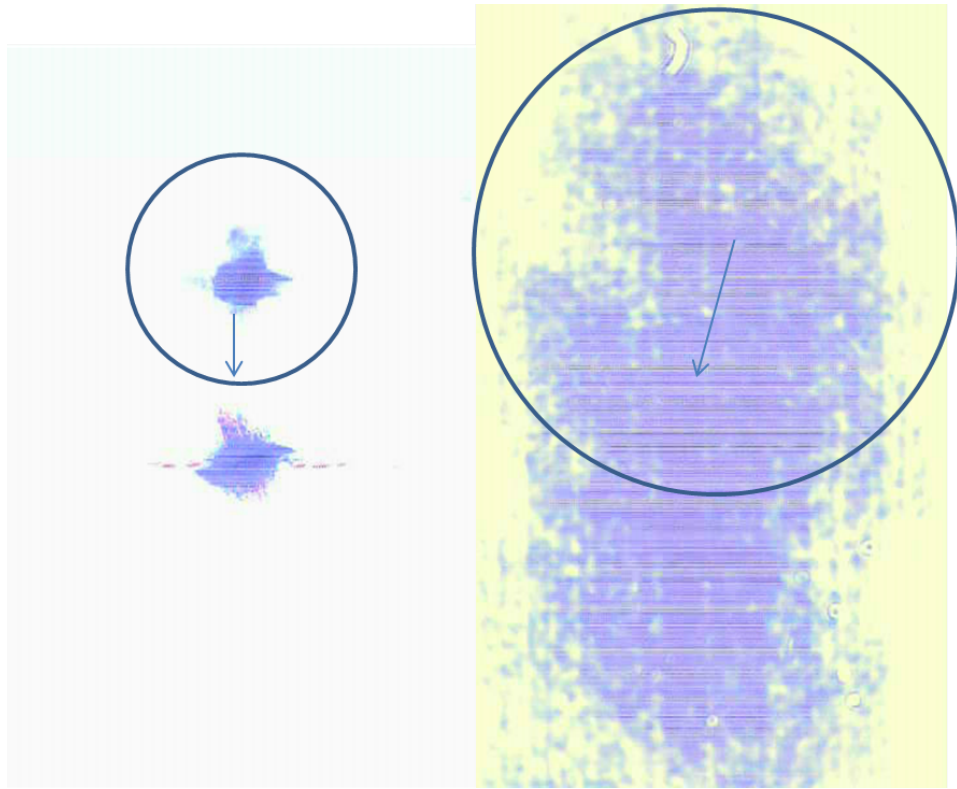


Fig. 4.5 Gate and test pulses overlap in the near and far field imaged with a camera put in(left) and out of focus (right)

Then the webcam was moved out of focus and the beams were overlapped again, as shown in the right side of Fig. 4.5 . By iterating this process it was possible to establish a sufficient spatial mode overlap. The crystal was then placed in the focal plane and its position was optimized by maximising the Type I SHG signal coming

from the pump beam. Once the position of the crystal was fixed, the crystal was rotated back to phasematch Type II SFG between the pump and the visible beam. In order to remove the residual pump and visible light, which being collinear were overlapped with the SFG signal, an UV bandpass filter, providing 80% transmission from 240 to 395 nm, was placed before the spectrometer.

4.3.3. RESULTS

Given the bandwidth of the SFG phase matching, and the UV bandpass filter, as well as the power of the pulses, by scanning 1mm path length with the stage on the gate arm (pump), it was possible to record simultaneously the traces of the XFROG between pump and pi, 2s and ps, as showing in Fig. 4.6.

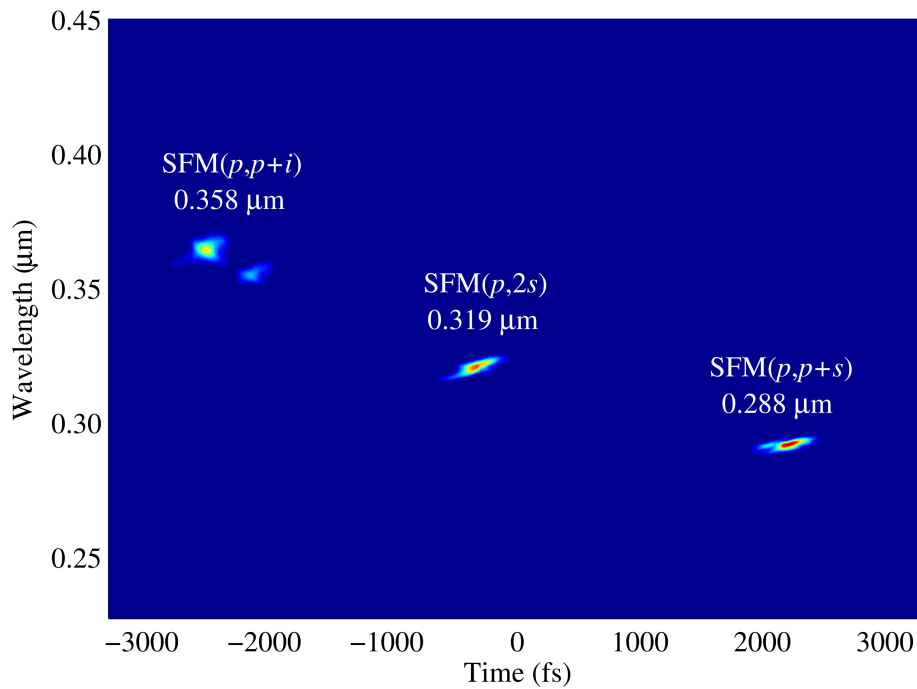


Fig. 4.6 Experimental XFROG traces for p+i, 2s and p+s pulses

It is immediate to notice from such a trace that there is a huge delay between the pulses. It needs to be noted that despite being collinear, there was a slight spatial

walk-off among the visible pulses. This resulted in the fact that it was impossible to optimize the overlap between the pump and all the visible pulses at the same time, and a compromise had to be made to acquire all the XFROG traces at the same time.

To retrieve the pulses, a standard PCGP algorithm was used. In order to remove the background signal given by the doubling from Type I SHG of the pump, individual traces of each arm were recorded and subtracted from the XFROG trace.

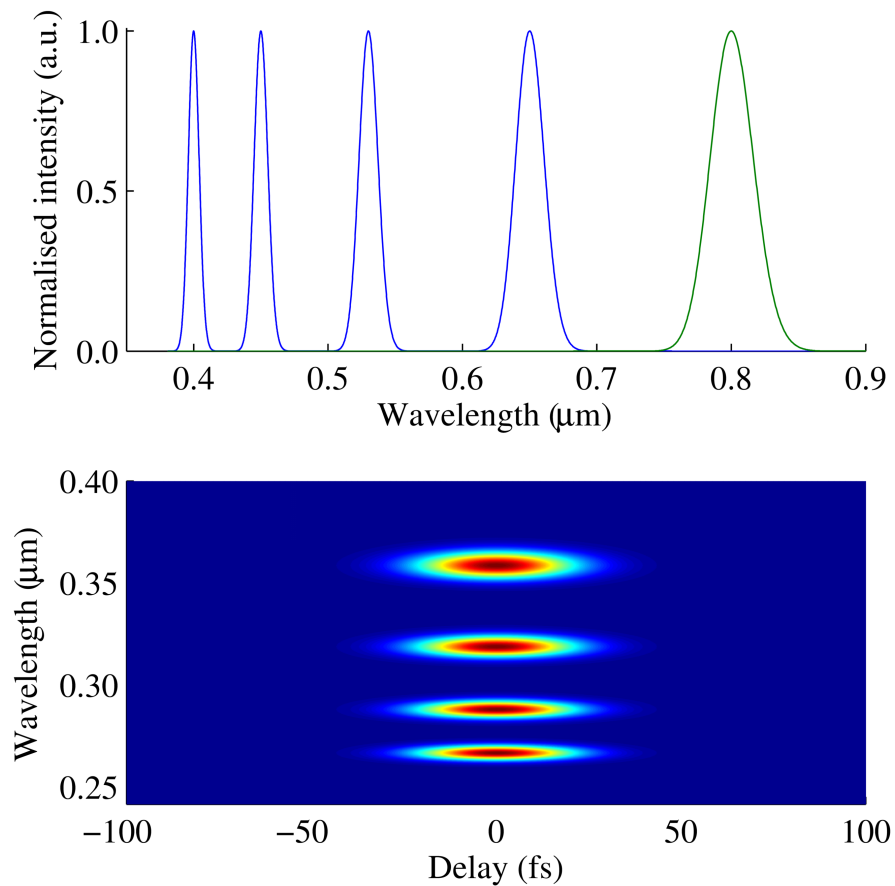


Fig. 4.7 Simulation of the XFROG trace (bottom) between the pump and the pulses in the top panel

The initial guess for the algorithm was given by the simulated FROG trace shown in Fig. 4.7. In 4.7 also the 400 nm pulse is reported for completeness, but its XFROG trace was not observed due to the limit of the UV bandpass filter.

In order to improve the algorithm robustness a constraint was imposed on the pump spectrum which was replaced in the algorithm with the spectrum measured experimentally. The retrieved spectra and duration of the pulses calculated after 40 iterations are shown in Fig. 4.8. The measured and retrieved spectra show good agreement suggesting a sensible reconstruction of the individual pulses.

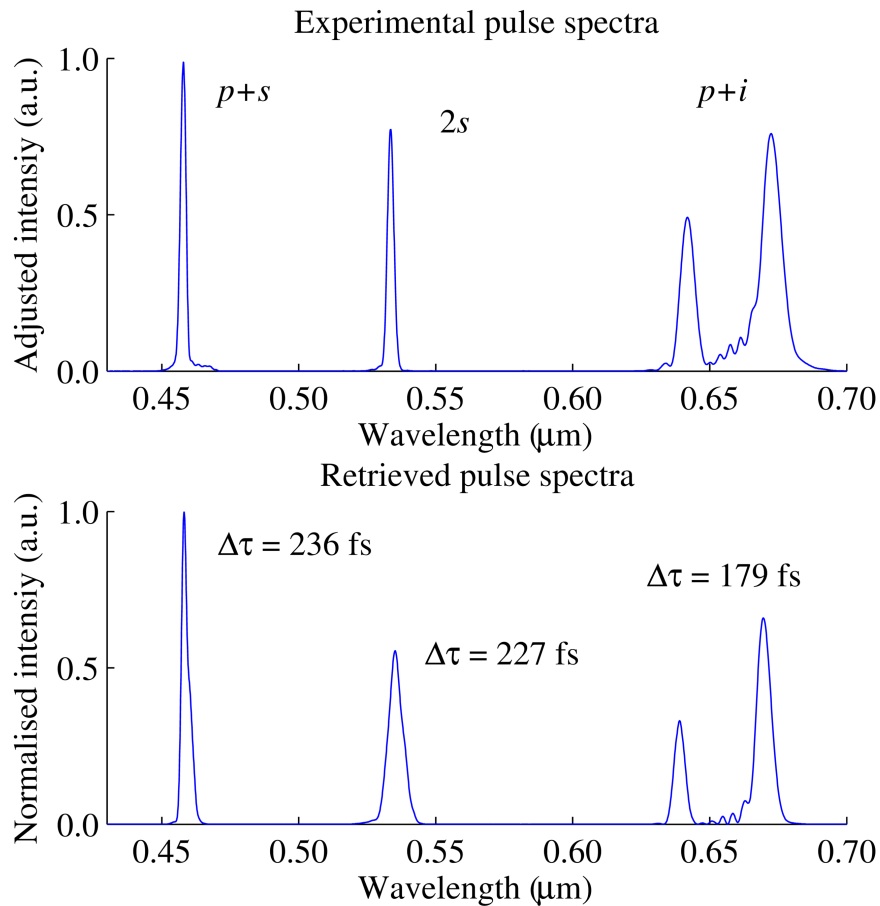


Fig. 4.8 Experimental and retrieved visible pulses spectra and FWHM duration calculated via the PCGP algorithm

As shown in Fig. 4.8 the FWHM of the p+i, 2s and ps pulses were respectively 236 fs, 227 fs and 179 fs which is far from the transform limited FWHM. This is due both to chirp introduced in the cavity and in the XFROG apparatus (as for example beam splitter cubes and HWPs). In the next section I will show how we then implemented the compression of the pulses to reach values close to the transform limited FWHM.

4.4. PULSE COMPRESSION USING A QUASI-COMMON-PATH PRISM DELAY LINE

The XFROG traces in Fig. 4.6 are useful to gather some important information about the pulses coming out of the OPO:

- the time of arrival of the OPO components are not equal, there is a very large GD between them;
- the individual traces of the OPO components are broadened indicating that the components are chirped.

In order to obtain the best synthesis possible these two factors need to be compensated. We decided to do so for three pulses in particular, the pump itself, which is a requirement for measuring all the other pulses, the 2s, and p+s components.

To use the XFROG to control the compression and group-delay compensation the setup of Fig. 4.9 was realised. Three selected frequencies from the OPO output are selected and sent to a quasi-common path (QCP) delay line. The beams were dispersed by a fused silica prism into three different delay lines where each was recollected by another fused silica prism which as we will see had been positioned to optimize the compression of the pulse. We chose to use prisms instead of gratings

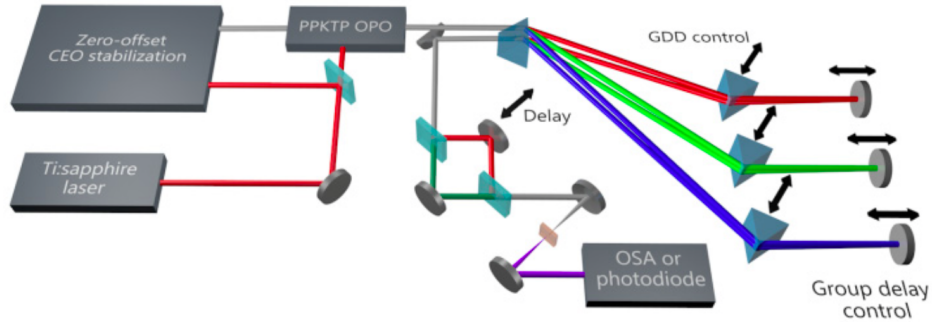


Fig. 4.9 Experimental setup for the compression and overlap of the OPO visible pulses

to minimize the losses and because of the large bandwidth required. The delay lines were then folded placing mirrors after the prisms and a vertical tilt was given to collect the output beam into the XFROG device.

4.4.1. PARENT PULSES COMPRESSION

In order to compress the pulses, individual XFROG traces for the three separate delay lines were acquired starting with the pump assuring the absence of any chirp bias when using then the pump as the reference for the XFROG of the other two delay lines. Starting from a prism difference of 38 cm the prism difference was varied to reduce the chirp which is clearly noticeable from the XFROG traces as it results in the broadening of the trace itself.

Fig. 4.10 shows the trace of the pump before being compressed, compared to Fig. 4.11 which is obtained for the optimized distance between the prism on the pump delay line which is found to be equal to 208 cm.

The trace obtained, despite having the same pulse as test and gate, was analysed with the XFROG algorithm instead of the SHG-FROG algorithm to take into account the fact that the arms were not entirely balanced. The results show that by changing

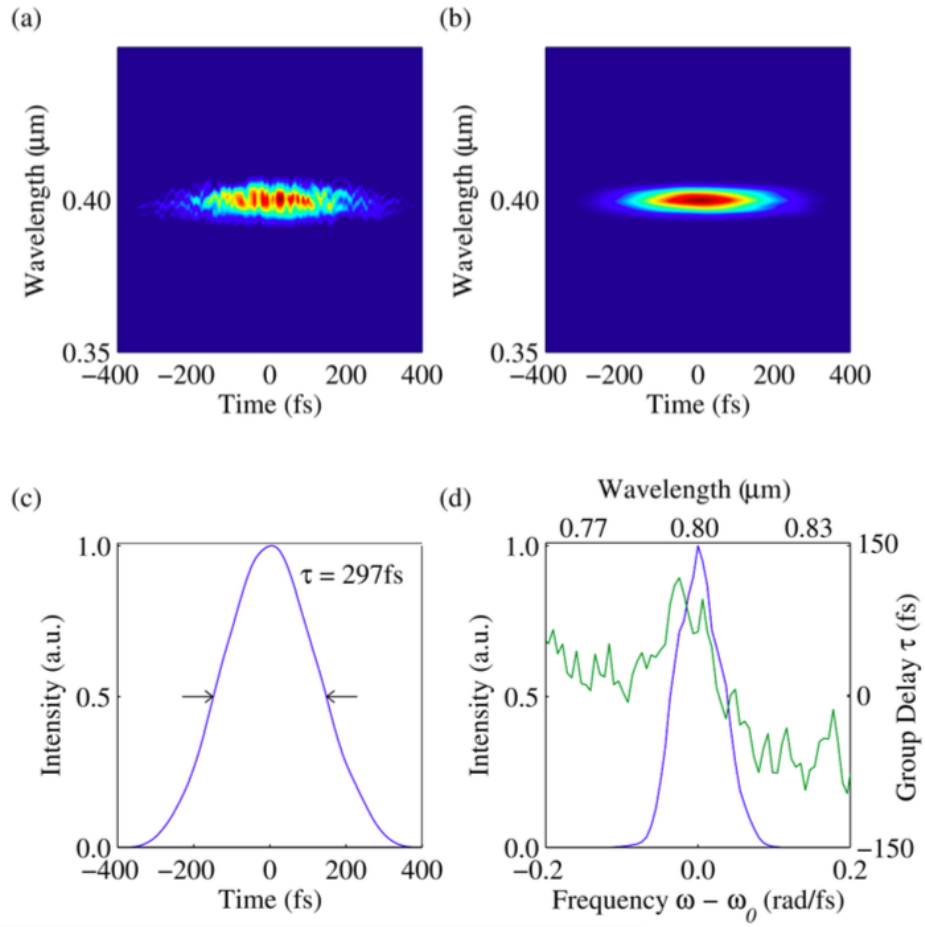


Fig. 4.10 Measured (a) and retrieved (b) pump-pump FROG trace before adjusting the delay line dispersion. (c) Time profile and (d) spectrum of the reconstructed pump pulse are shown. The pulse exhibits a FWHM of $\tau = 297\text{ fs}$.

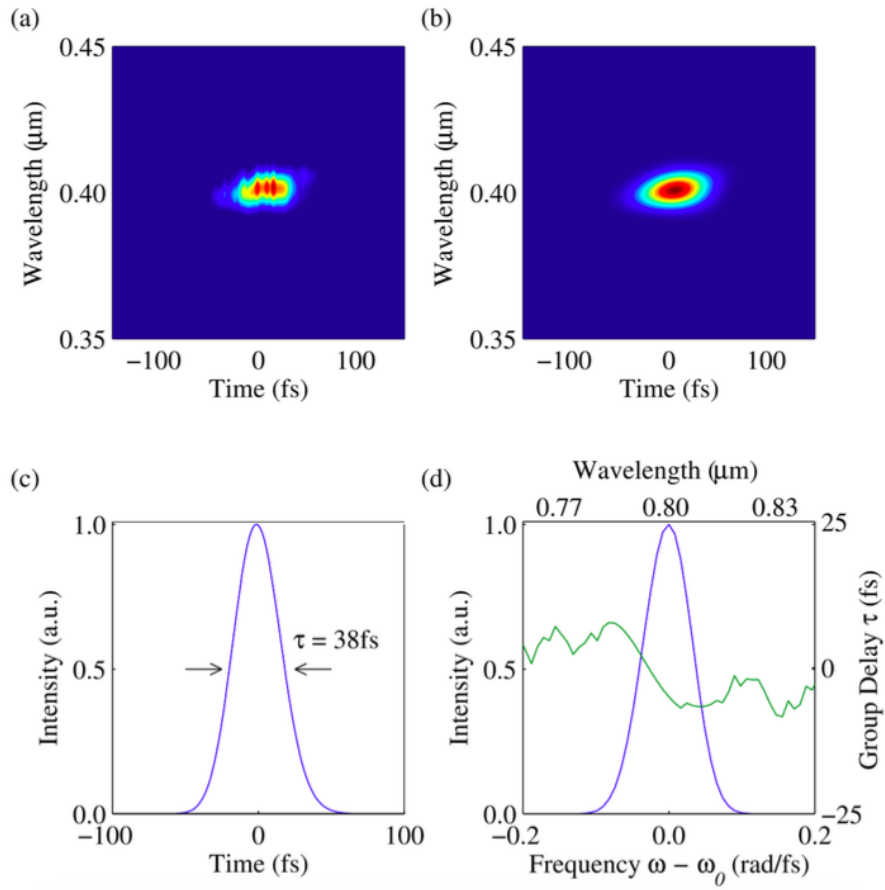


Fig. 4.11 Measured (a) and retrieved (b) pump-pump FROG trace after adjusting the delay line dispersion. (c) Time profile and (d) spectrum of the reconstructed pump pulse are shown. The pulse exhibits a FWHM of $\tau = 38\text{ fs}$.

the prism distance it was possible to compress the pulse from $\Delta\tau_{FWHM} = 297$ fs to $\Delta\tau_{FWHM} = 38$ fs, compared to a transform limited duration of 37 fs.

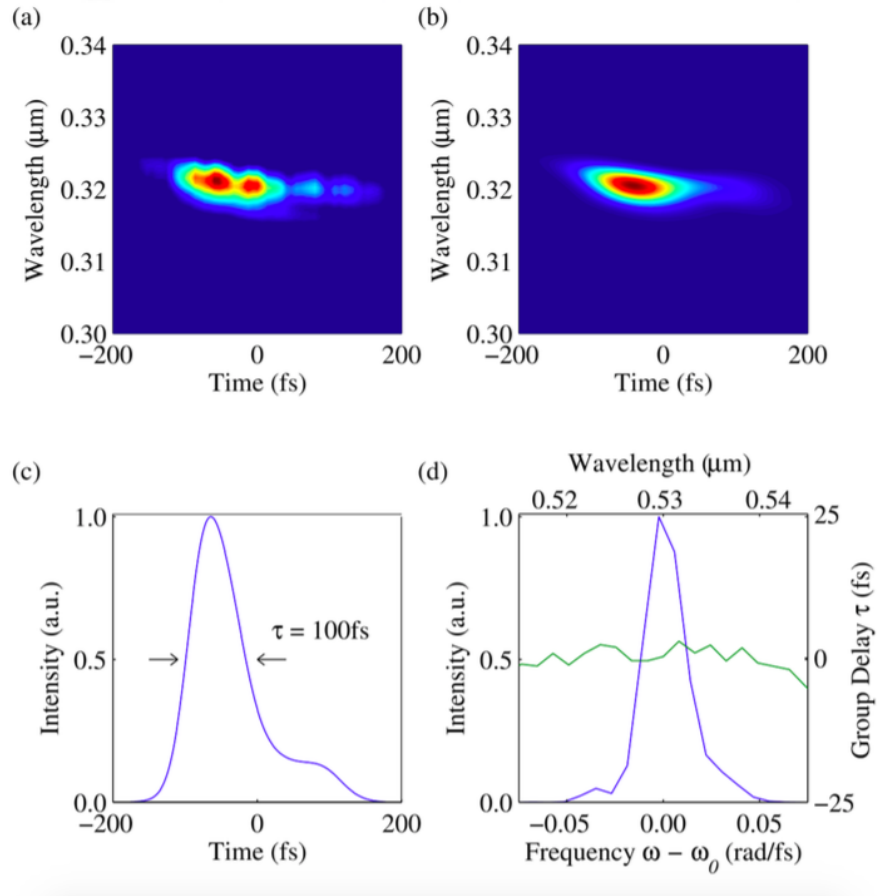


Fig. 4.12 Measured (a) and retrieved (b) pump-signal (2s) XFROG trace after adjusting the delay line dispersion. (c) Time profile and (d) spectrum of the reconstructed pulse are shown. The pulse exhibits a FWHM of $\tau = 100$ fs

Once the pump compression had been optimized, it was possible to determine the compression of the other two pulses with an XFROG measurement using the pump as the reference. The results showed that for the double-signal pulse the optimal compression was obtained with a prisms distance of 150 cm and for the pump-signal pulse with a distance of 120 cm. The pulses were compressed respectively to 100

fs and 115 fs, indicating that they were both still slightly chirped, the transform limited duration being respectively 80 fs and 86 fs.

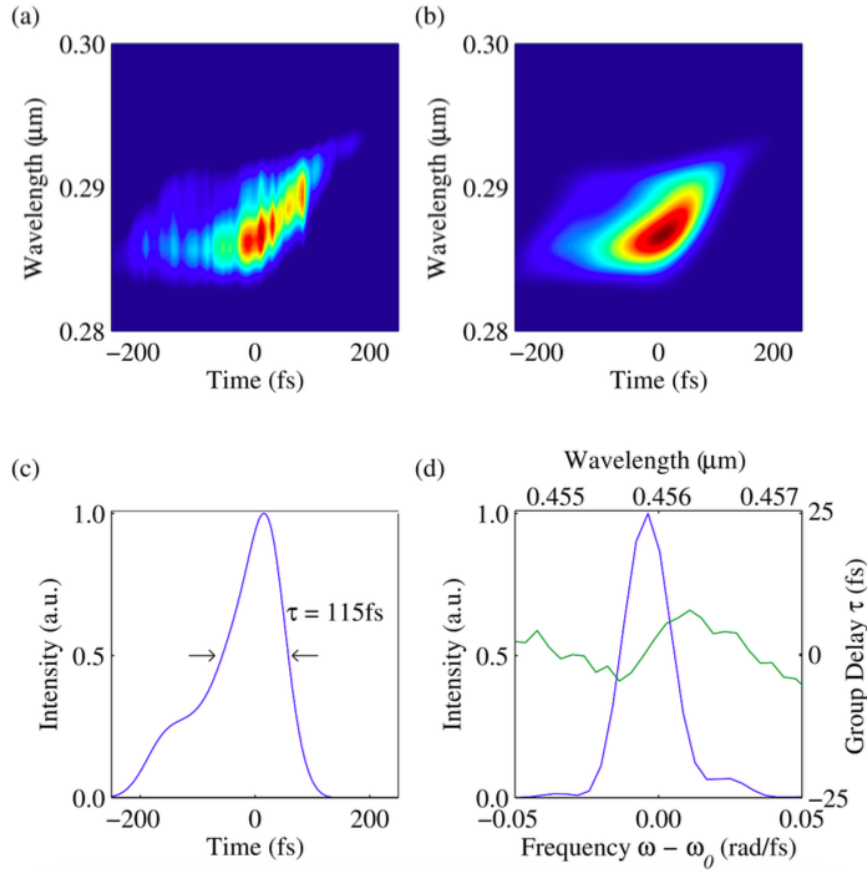


Fig. 4.13 Measured (a) and retrieved (b) pump - pump+signal (p+s) XFROG trace after adjusting the delay line dispersion. (c) Time profile and (d) spectrum of the reconstructed pulse are shown. The pulse exhibits a FWHM of $\tau = 115\text{fs}$

4.4.2. RELATIVE DELAY MANIPULATION

The other feature visible from the XFROG trace is that the pulses are not temporally overlapped, but they arrive with a relative delay on the spectrometer with respect to the pump. A simulate trace is shown in Fig. 4.14 in the case of pulses with relative delays (top) and zero relative delay (bottom).

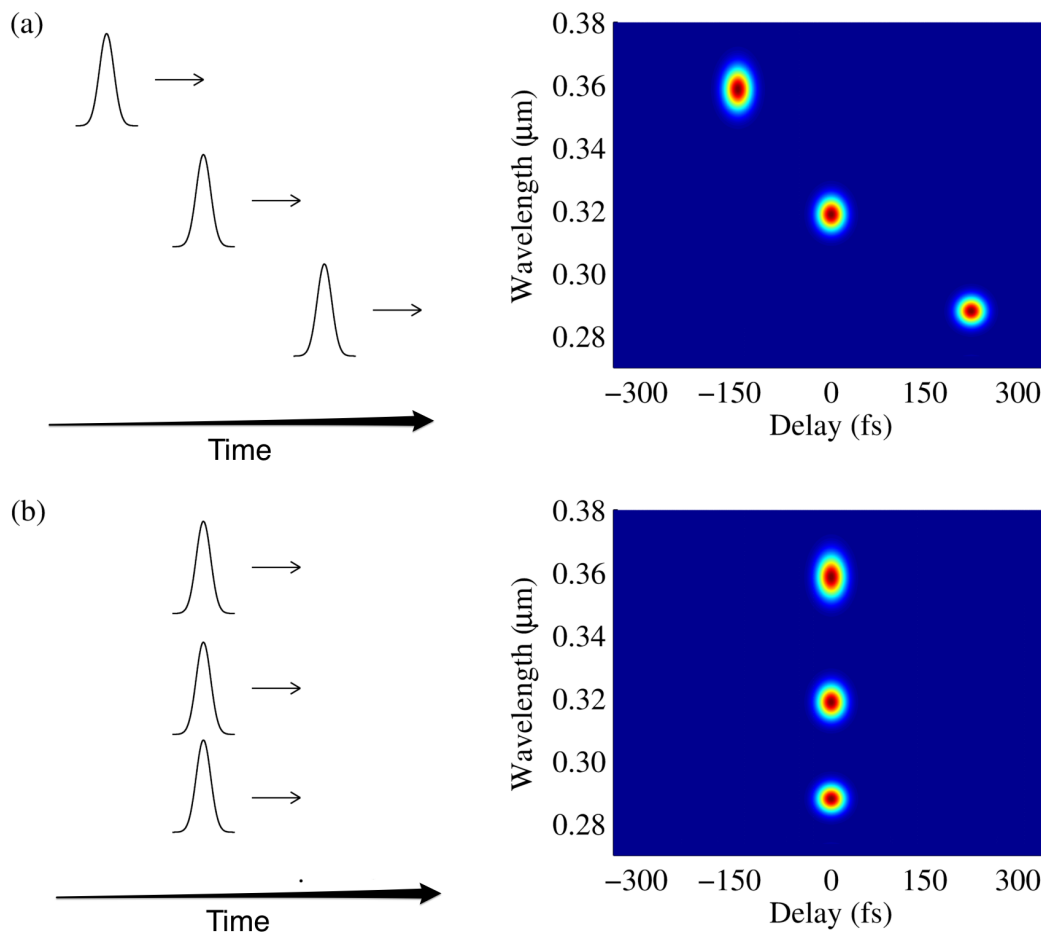


Fig. 4.14 Simulation of relative delay correction for three compressed parent pulses. in a) three simulated pulses with relative delay are reported together with their FROG trace. in b) the same pulses are shown when their relative delay is set to zero

To correct for this behaviour the temporal walk-off was modified by adjusting the positions of the folding mirror on the delay line of the $2s$ and ps pulses. The folding mirror position was first obtained by making the three path lengths equal with a measuring tape and then adjusted with a micrometre stage.

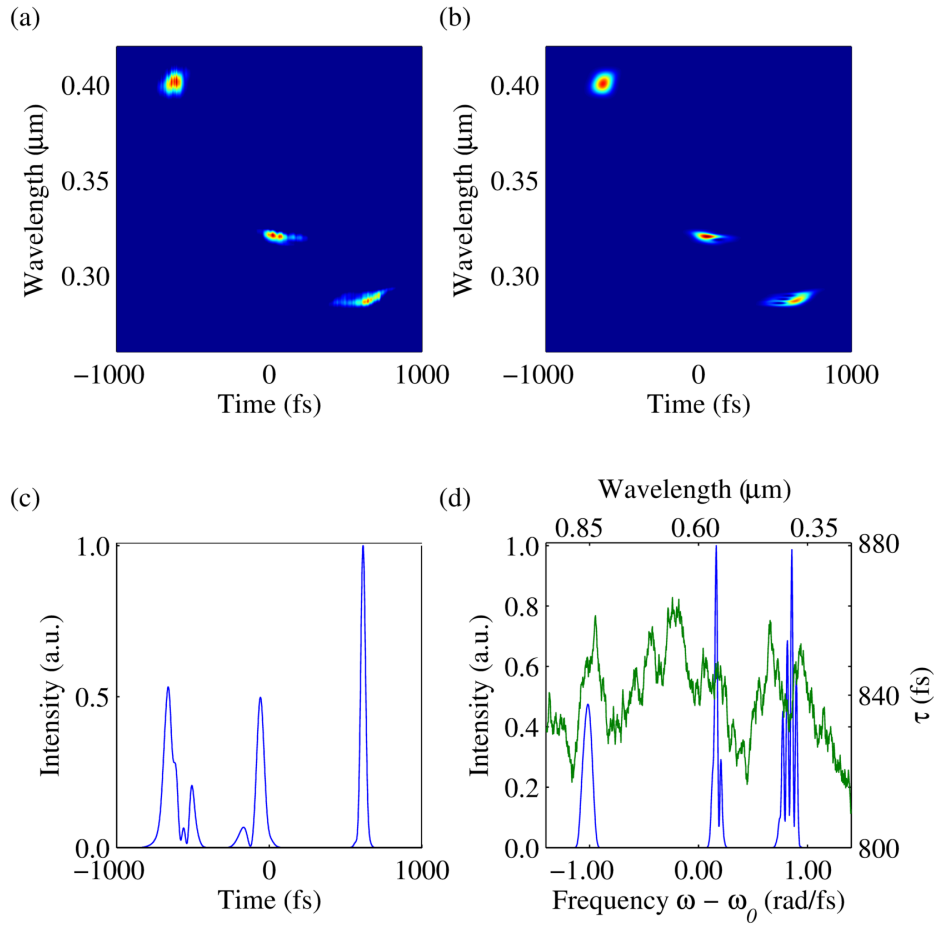


Fig. 4.15 Results of the XFROG measurements on pump, doubled-signal and pump+signal pulses before optimizing the quasi-common path delay lines.

In Fig. 4.15 the results of the XFROG before the micrometre adjustment are reported. The delay between the leading and trailing pulses amounts to approximately 1.5 ps. This delay, equal to about 0.5 mm of air propagation was then corrected by iteratively acquiring the XFROG trace and moving the mirror stage

position accordingly. The results obtained after the adjustment are shown in Fig. 4.16. From the panels (c) both in Fig 4.15 and 4.16 which show the reconstructed pulse in the time domain it is clear how the pulses went from being temporally separated to temporally overlapped.

It must be however noted that, as previously said, the XFROG method fails to completely characterize the pulse and nothing can be said, from the XFROG traces analyzed, about the OPO components' relative phases. Thus, even if each of the individual pulses has been characterized nothing can be conclusively said about the coherently-synthesised pulse resulting from their overlap. The results discussed are only an estimate obtained by setting the relative phases of the pulses to zero.

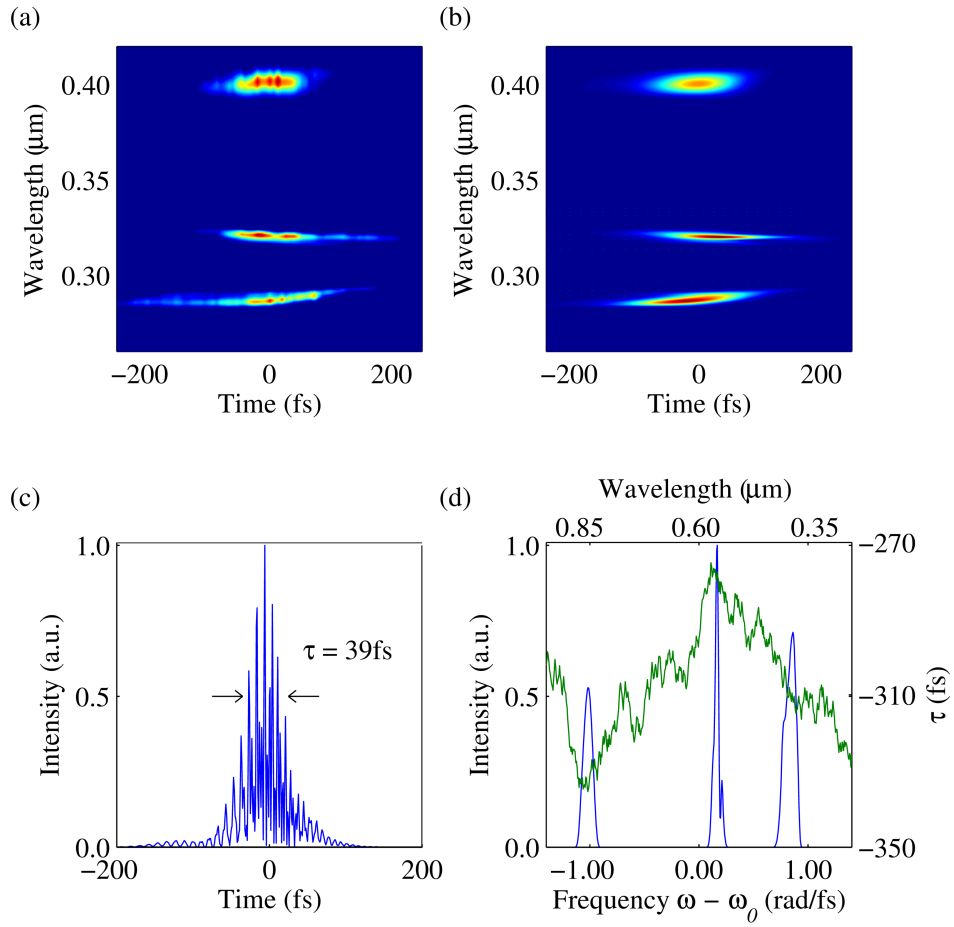


Fig. 4.16 Results of the XFROG measurements on pump, doubled-signal and pump+signal pulses after the optimization of the quasi-common path delay lines.

CHAPTER 5

SPECTRAL-GAP IMMUNE CHARACTERIZATION OF ELECTRIC FIELDS

"He who controls the Spice, controls the universe"

- Dune, 1984 -

5.1. INTRODUCTION

In this chapter I will discuss the main result of my thesis: a new interferometric technique for the characterization of pulses with spectral gaps. The technique, named SPectral-gap Immune Characterization of Electric-fields (SPICE)¹, comes from the entanglement of the SPIDER - and in particular the SEA-CAR implementation, and the MICE techniques. By combining the multi-shear approach typical of the SEA-CAR SPIDER and the multiple fields reconstruction delivered by MICE, it allows the use of an unknown reference to carry the information of the relative

¹No intended reference to fictional literary sagas, movies featuring Sting, nor 90s pop groups.

phase across the spectral gap, It also allows for the characterization of the third field which causes the failure of the SEA-CAR SPIDER approach. After describing the method and the algorithm developed through different simulation examples, I will discuss the experiment I have implemented and the results obtained.

5.2. THE SPICE METHOD

In order to describe the method behind this new technique, I will refer to the simplest case in which there is only one spectral gap to be bridged, resulting in two well-separated spectral features as in Fig. 5.1.

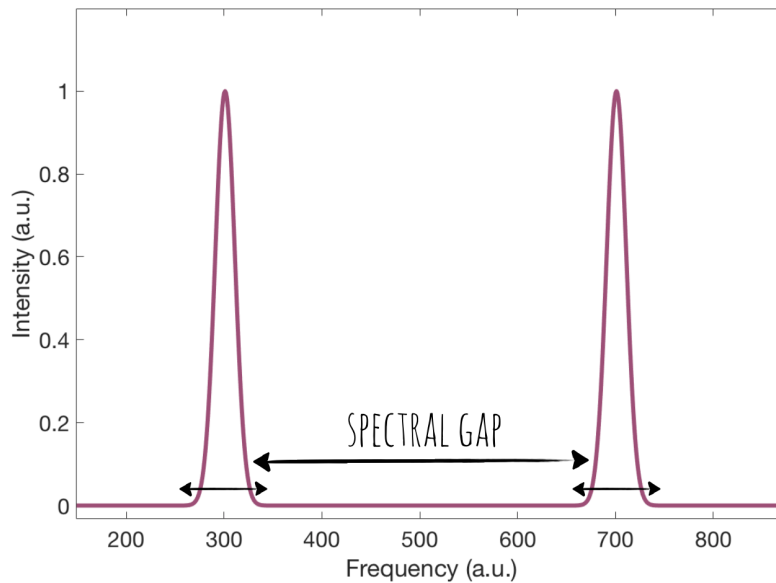


Fig. 5.1 Example of a pulse which exhibits a *spectral gap*

The challenge is thus to reconstruct the relative phase between the two distinct spectral features and to do so there is need to find an original way to transfer the phase information across the spectral null trying to retain the higher number of features from previous techniques, as for example having single-shot measurements,

avoid the need of a known reference, and to keep a manageable experimental setup. The SPICE method, can be summarised in three steps:

1. preparing the pulse under study so that the gap can be bridged;
2. making the prepared pulse interfere in a way that the gap phase information is not destroyed;
3. applying an appropriate algorithm to extract the information required.

FIRST STEP: SIGNAL PULSE PREPARATION

The first step of the SPICE protocol consists in preparing the test pulse (TP) so that the information can be carried along the spectral intensity nulls. To do so, based on existing technique it is straightforward to imagine adopting a multi-shear approach, with the minimum shear small enough to guarantee a sufficient resolution for the phase reconstruction and the maximum shear large enough to shift the first spectral peak to overlap with the second spectral peak. The shear can be applied either temporally (as it happens in SPIDER) or spatially (as in SEA-CAR-SPIDER) but adopting the former would mean to have consecutive measurements for different shears abandoning the single-shot capabilities which can be required for some experiments, thus the spatial shear approach is chosen.

This is achieved by upconversion of the TP with a spatially chirped ancillary pulse (AP), as shown in Fig. 5.2. The spatial chirp is arranged so that the spatial coordinate x and the shear Ω have a simple linear relation: $\Omega = \alpha \cdot x$, where the constant α can be obtained via calibration.

An essential requirement of the AP is that its bandwidth is large enough to cover the separation between the two TP peaks. If that is the case, there will exist at least two given positions (i.e. two values of the shear) in which the two spectral

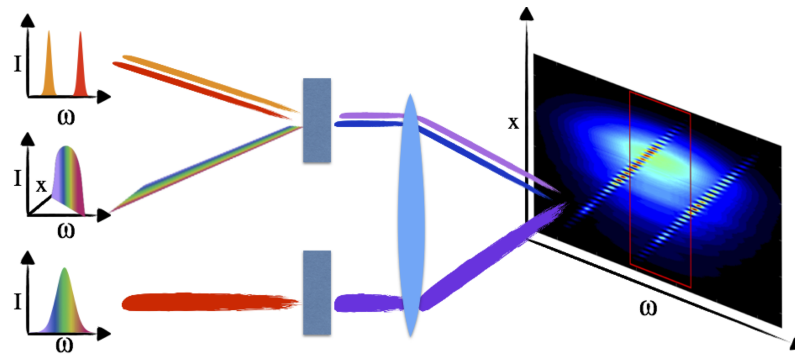


Fig. 5.2 Spatial shear scheme

peaks are centred at the same frequency, as it happens for all the frequencies value of the so obtained signal pulse (SP) in the purple rectangle in Fig. 5.3.

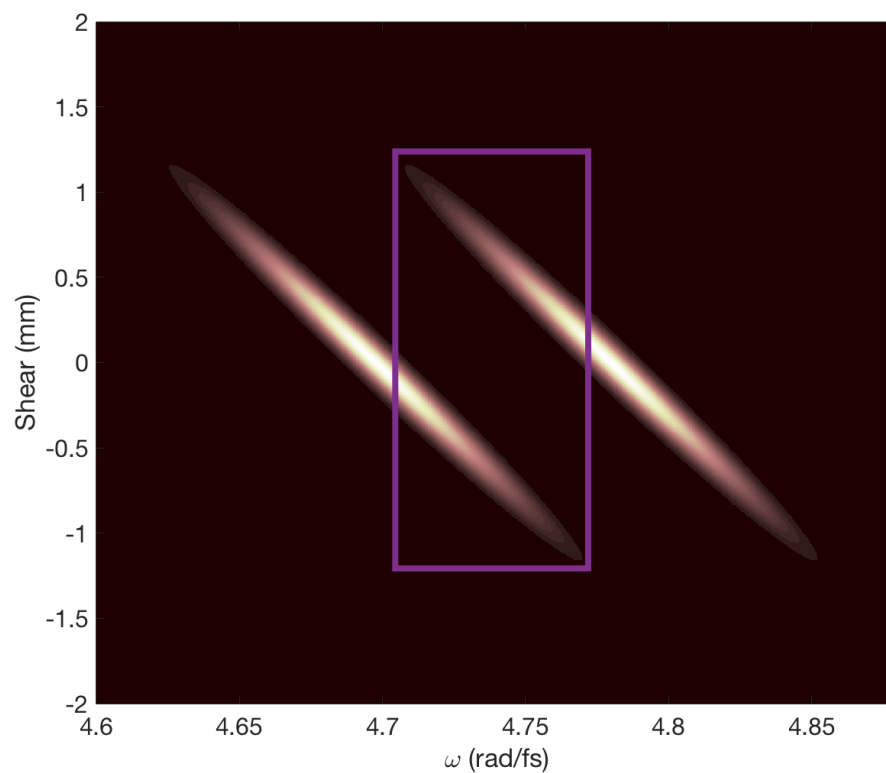


Fig. 5.3 Spatially chirped SP

Using the upconversion for obtaining the shear is very common in many metrology techniques, as discussed in Chapter 3; however it must be noted that by doing so an additional phase is introduced depending exclusively on the shear Ω , $\Phi_{AP}(\Omega)$. This is due to the fact that the AP has a phase which is not necessarily flat, and thus since the spectrum of the ancilla is mapped in space because of the spatial shear of the AP, Φ_{AP} is constant for each position, but varies along the spatial axis, that is, with the shear. As it will be explained in the third step, this will be crucial for the choice of the retrieval algorithm.

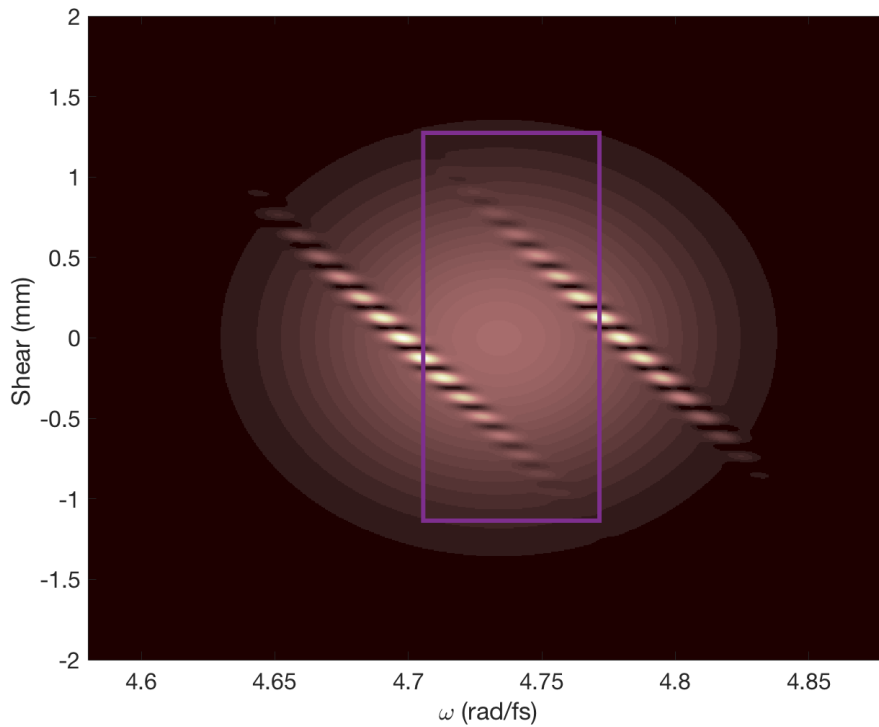


Fig. 5.4 Example of a SPICE interferogram for a TP with a spectral gap.

SECOND STEP: INTERFERENCE

In the second step the spatially sheared SP is made interfere either spatially or spectrally with a reference pulse (RP). The phase of the RP can be unknown. The

only requirement for the AP is that it must have a spectrum with sufficient bandwidth to cover the SP's entire spectrum. The interferogram observed is recorded by means of an imaging spectrometer.

The interference pattern between the SP and the RP reads:

$$I(\omega, \Omega, x) = |\mathcal{E}_{RP}(\omega) \cdot a_{RP}(x)|^2 + |\mathcal{E}_{SP}(\omega - \Omega) \cdot a_{SP}(x)|^2 + \dots \quad (5.1)$$

$$\dots + 2\text{Re}(\mathcal{E}_{RP}(\omega) \cdot \mathcal{E}_{SP}^*(\omega - \Omega) \cdot \mathcal{A}(x)),$$

where $\mathcal{E}_i$ and a_i are the frequency and spatially dependant components of the complex electric fields of pulse i and $\mathcal{A}(x) = a_{RP}(x) \cdot a_{SP}(x)$. An example of such interferogram is given in Fig. 5.4

THIRD STEP: RETRIEVAL ALGORITHM

We now have to extract the phase information from the interferogram of Eq (5.1). We can notice that for any frequency ω_0 in the highlighted region in Fig. 5.4 we can isolate two interferograms, each associated with a different spectral peak sheared by an amount $x_1(\Omega_1)$ and $x_2(\Omega_2)$ respectively. In this frequency range it is meaningful to compare the interferometric phases, given by the argument of the oscillatory term in Eq. (5.1):

$$\Gamma_1(\omega_0) = \phi_{SP1}(\omega_0) - \phi_{RP}(\omega_0) - \Phi_{AP}(\Omega_1) \quad (5.2)$$

$$\Gamma_2(\omega_0) = \phi_{SP2}(\omega_0) - \phi_{RP}(\omega_0) - \Phi_{AP}(\Omega_2),$$

where ϕ_{SP1} and ϕ_{SP2} are the phases of the spectral peaks within the SP. If $\Phi_{AP}(\Omega_1) = \Phi_{AP}(\Omega_2) = 0$ held true, it would be trivial to obtain the relative phase between the two spectral peaks by taking the difference of the two measurements, and the spectral phase could be retrieved by adopting a standard SPIDER concatenation

algorithm. However, as we have discussed in Step 1, the phase varying with the shear, which is given by the phase of the Ancillary pulse, is not necessarily flat and equal to zero. Since this is the case, it is no longer straightforward to retrieve the phases from Eq. (5.2) as too many unknown fields are present. This is why there is need of a new algorithm which can reconstruct multiple fields at the same time, which, as described in Chapter 3, is one of the main feature of the MICE algorithm. The MICE equations for the three fields TP, AP and RP, as derived in Chapter 3, are:

$$\begin{aligned}
 \mathcal{E}_{RP}(\omega_j) &= \frac{\sum_k AC_{j,j-k}^{meas} \cdot \mathcal{E}_{TP}(\omega_j - \Omega_k) \cdot \mathcal{A}^*(\Omega_k)}{\sum_k |\mathcal{E}_{TP}(\omega_j - \Omega_k) \cdot \mathcal{A}(\Omega_k)|^2} \\
 \mathcal{E}_{TP}^*(\omega_j) &= \frac{\sum_k AC_{j+k,j}^{meas} \cdot \mathcal{E}_{RP}(\omega_j + \Omega_k) \cdot \mathcal{A}^*(\Omega_k)}{\sum_k |\mathcal{E}_{RP}(\omega_j + \Omega_k) \cdot \mathcal{A}(\Omega_k)|^2} \\
 \mathcal{A}(\Omega_k) &= \frac{\sum_j AC_{j,j-k}^{meas} \cdot \mathcal{E}_{TP}^*(\omega_j - \Omega_k) \mathcal{E}_{RP}(\omega_j)}{\sum_j |\mathcal{E}_{TP}(\omega_j - \Omega_k) \mathcal{E}_{RP}(\omega_j)|^2}.
 \end{aligned} \tag{5.3}$$

Eq. (5.3) can be solved using an iterative algorithm which minimise the MICE error:

$$Err = \sum_{j,k} |AC_{j,k} - \mathcal{E}_{TP}(\omega_j - \Omega_k) \cdot \mathcal{E}_{RP}(\omega_j) \cdot \mathcal{A}(\Omega_k)|^2, \tag{5.4}$$

where $AC_{j,k}$ is the measured AC band and the other term is the reconstructed one. Solving Eq. (5.3) enables the simultaneous reconstruction of the TP, AP and the RP, hence overcoming the difficulties of having an additional shear-varying ancillary phase. Using the MICE algorithm to extract the phase has many perks: firstly due to the redundancy of the multi-shear approach, the algorithm is highly efficient and very solid to noise perturbations. Furthermore, it allows exploiting of the benefits coming from an external source for power and bandwidth without adding to the TP reconstruction any additional errors coming from a characterization of the reference

itself, as there is no need to know any information about the reference apart that it has a suitable spectrum and power, which are both easy conditions to check. It is also worth noting that there is no need for the algorithm to characterize the whole spectral phase of the RP, in the eventuality that the RP's spectrum is larger than the SP's: the RP phase will in fact be reconstructed only in the spectral range which interferes with the SP, and that is all that is required for the algorithm to provide the SP characterization.

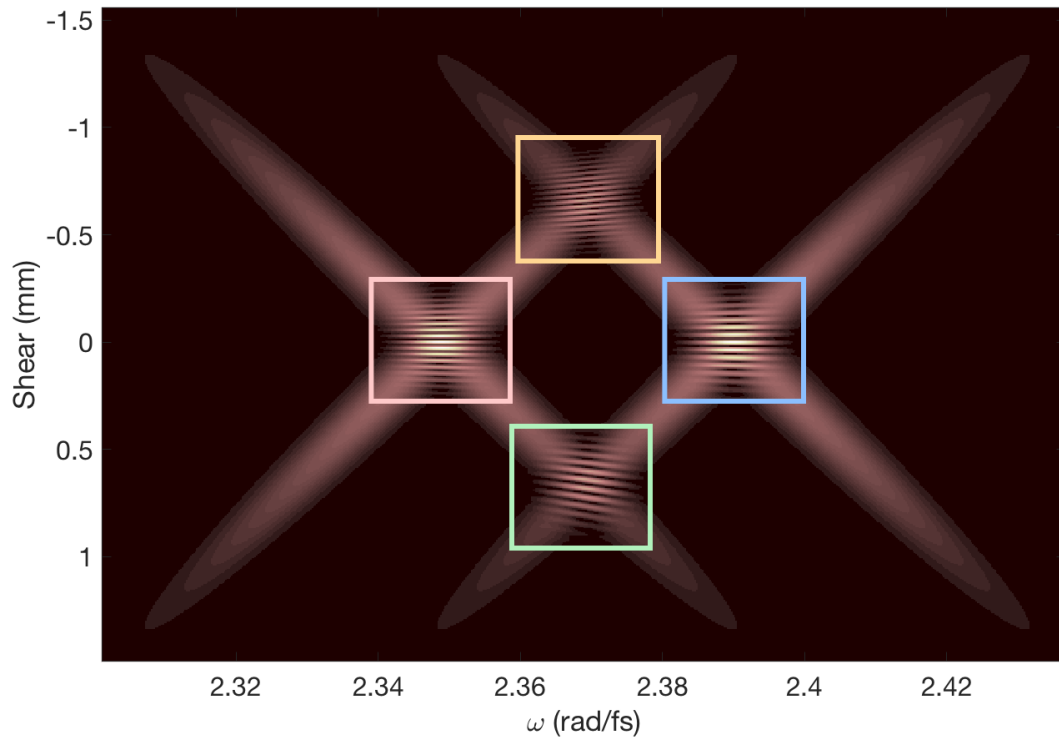


Fig. 5.5 Example of a SEA CAR SPIDER interferogram on a gapped pulse

5.2.1. COMPARISON WITH THE SEA-CAR SPIDER APPROACH

The SEA-CAR SPIDER technique suffers from an intrinsic limitation when it comes to spectral gaps: The shear-varying phase $\phi_{AP}(\Omega)$, which arises from the TP upconversion – that is, the very method used to shear the test pulse – adds a

variable to the reconstruction making it impossible to relate directly the information on two different spatial points of the interferogram.

Let's consider the example of Fig. 5.5 where a SEA CAR SPIDER interferogram is simulated for a spectrally gapped pulse. If we label the spectral peaks of the two TP replicas $A_{1,2}$ for the first peak and $B_{1,2}$ for the second, we can identify four distinct interferograms areas as follows:

- Interferogram 1 (pink square): interference between peaks A_1 and A_2 ;
- Interferogram 2 (blue square): interference between peaks B_1 and B_2 ;
- Interferogram 3 (yellow square): interference between peaks A_1 and B_2 ;
- Interferogram 4 (green square): interference between peaks B_1 and A_2 .

From the interferogram 1 and 2 it is possible using either a standard SPIDER or MICE algorithm to retrieve the phase of the two separate spectral peaks. Interferogram 3 and 4 both provide information on the relative phase of the peaks as the interferogram phase reads: $\Phi_3 = \phi_{A,1}(\omega + \Omega) + \phi_{B,2}(\omega - \Omega) + \phi(\Omega)$ for Interferogram 3 and $\Phi_4 = \phi_{B,1}(\omega + \Omega) + \phi_{A,2}(\omega - \Omega) + \phi_{AP}(\Omega)$ for Interferogram 4. Failing to reconstruct the phase $\phi_{AP}(\Omega)$, a standard SPIDER approach can not thus provide any information on the relative phase of the two peaks. Using MICE the phase $\phi(\Omega)$ can be also retrieved but it should be noted however that this is only possible *locally*: It is possible to reconstruct it for the shear values in interferogram 1,2,3,4 but since those are not linked, it is impossible, by solely using MICE, to retrieve any information on the relative phase. This is instead solved by using the SPICE technique which, by means of a broad unknown reference guarantees the reconstruction of $\phi(\Omega)$ along the whole region of interest.

5.3. SIMULATIONS

In order to check the consistency of the algorithm I have performed simulations firstly to verify the known results when an unfiltered TP was considered, then to verify the relative phase reconstruction with a filtered TP, and furthermore to study the persistence to noise of both the unfiltered and filtered TP reconstruction. In order to do so, uniformly distributed noise is added to each pixel of the interferogram and the noise level is given by a random number from the interval (0,1) times a percentage of the maximum intensity of the interferogram. As it will be shortly illustrated, and had been already pointed out in [96] the algorithm also in this instance appears very robust to noise.

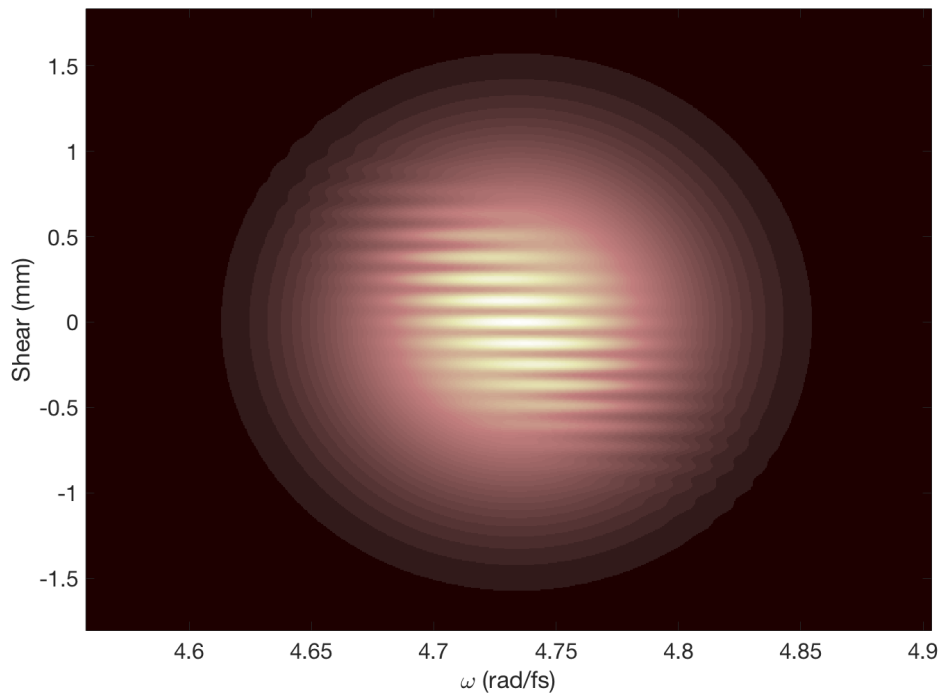


Fig. 5.6 Simulated SPICE interferogram

5.3.1. UNFILTERED TP SIMULATION

Using the SPICE algorithm I performed a simulation to reconstruct the phase of a Gaussian pulse without spectral gaps. This is a standard result which can be obtained with the most simple reconstruction algorithm but it serves here to verify that not only the phase of the TP is reconstructed but also those of the RP and AP are. The starting point of the simulation is artificially creating three Gaussian pulses for the TP, AP, and RP and their phases.

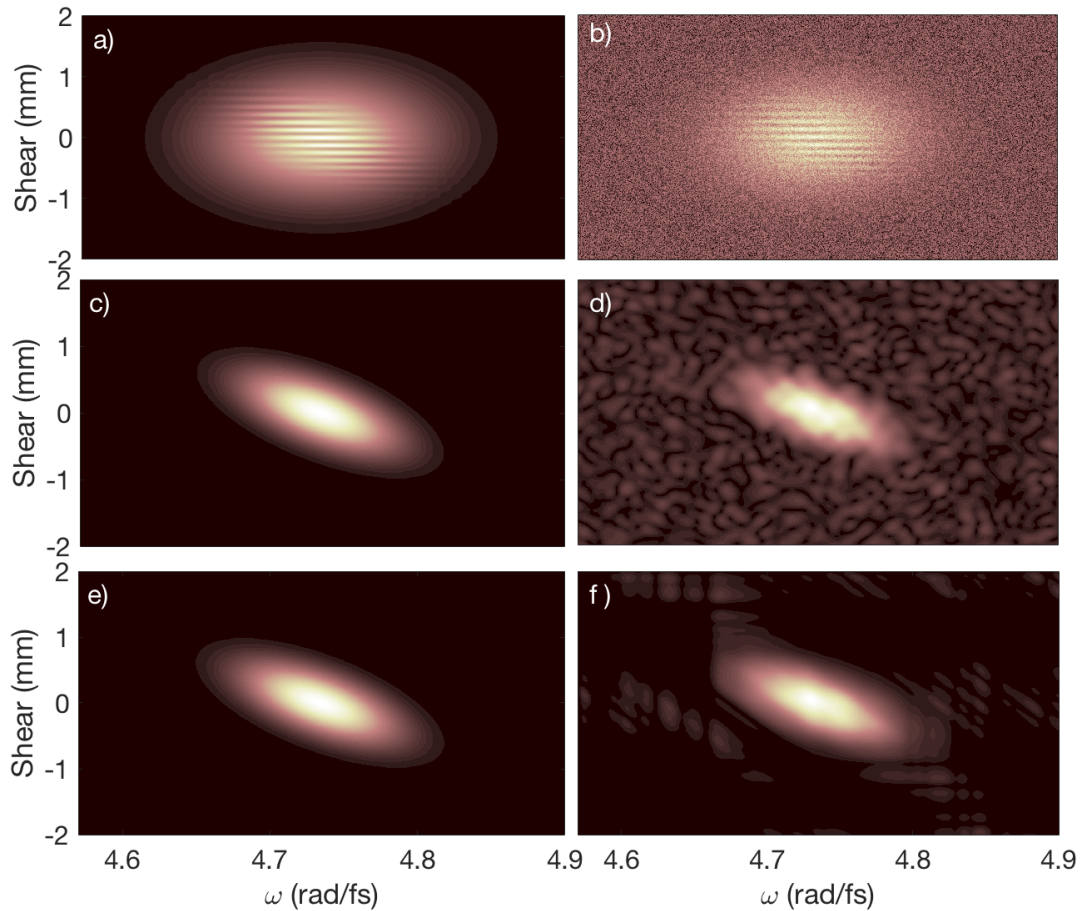


Fig. 5.7 Left column: interferogram (a), AC side band (c), retrieved AC side band (e) in the absence of noise; Right column interferogram (b), AC side band (d), retrieved AC side band (f) with 50% additive noise

The frequency and shear scale have been chosen to be similar to those expected in the experiment. For the example shown the phases of the TP and RP are chosen to be quadratic with coefficients: $GDD = 400 \text{ fs}^2/\text{rad}$ and the phase for the AP is $GDD = 4 \text{ rad/mm}^2$ (the different units are due to the fact that the AP in the simulation is constructed on the spatial shear axis, which is in mm, but as explained in the previous section the AP spectrum is mapped in the spatial dimension, so the two domains are equivalent). In Fig. 5.6 the SPICE interferogram obtained with such pulses is shown.

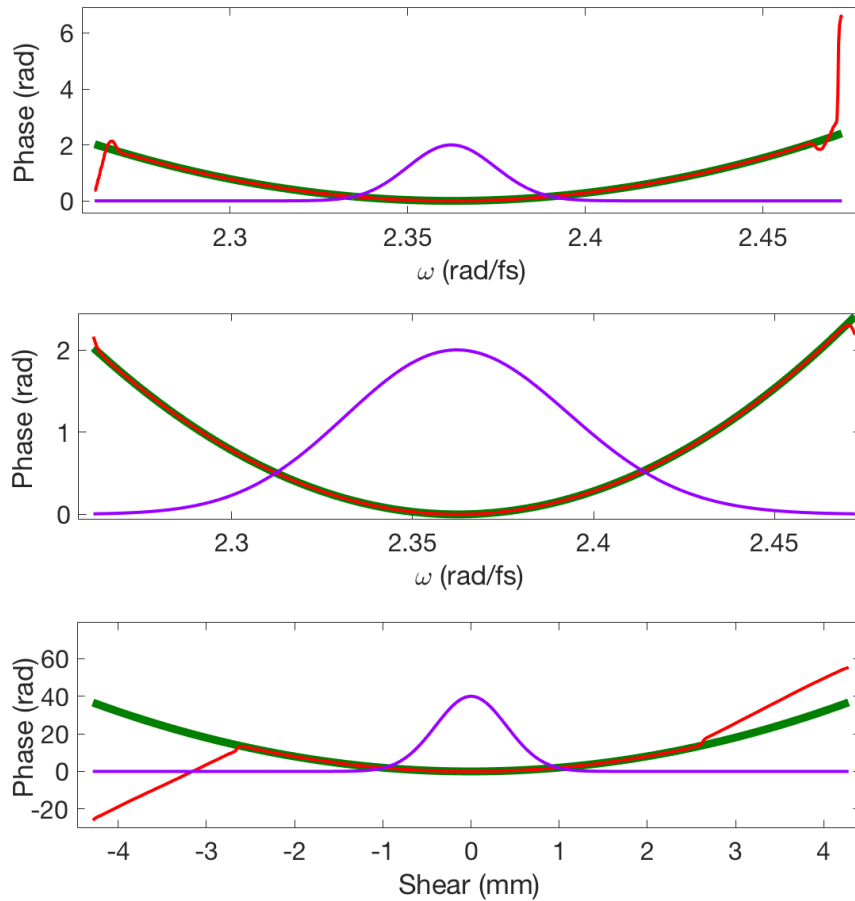


Fig. 5.8 Theoretical (green) phase , reconstructed (red) phase, and spectrum (purple) for the TP (top panel) RP (middle panel) AP (lower panel) in the case of zero noise

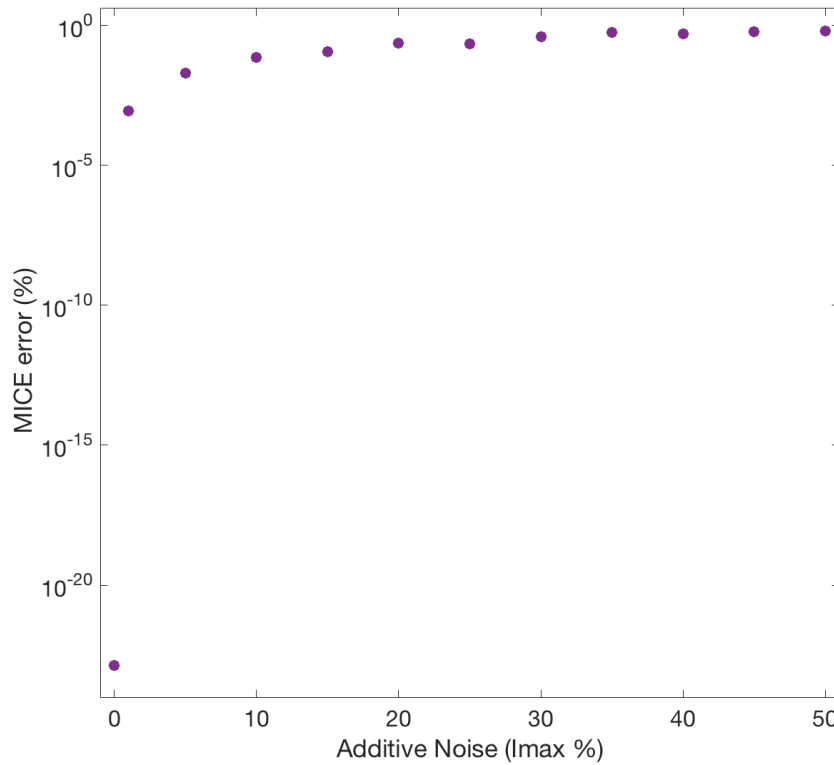


Fig. 5.9 Semi-log plot of the MICE error versus of the additive noise level.

A 2 dimensional FFT is then applied to the interferogram to isolate the sideband by using a filter in the Fourier domain, which is then Fourier-transformed back and used by the algorithm to retrieve the three parent fields according to Eq. 5.3. In Fig 5.7 the retrieved (c-d) and reconstructed (e-f) AC bands are shown together with the respective interferogram (a-b) referring to the case of zero noise and the case of 50% additive noise. Already from the reconstructed images it is possible to see that even in the extremely unfortunate scenario where the noise is 50% of the signal maximum, the AC band is satisfactorily reconstructed.

In Fig 5.8 the reconstructed and theoretical phases of the TP, REF and AP pulses are shown together with their spectra in the case where no additive noise is

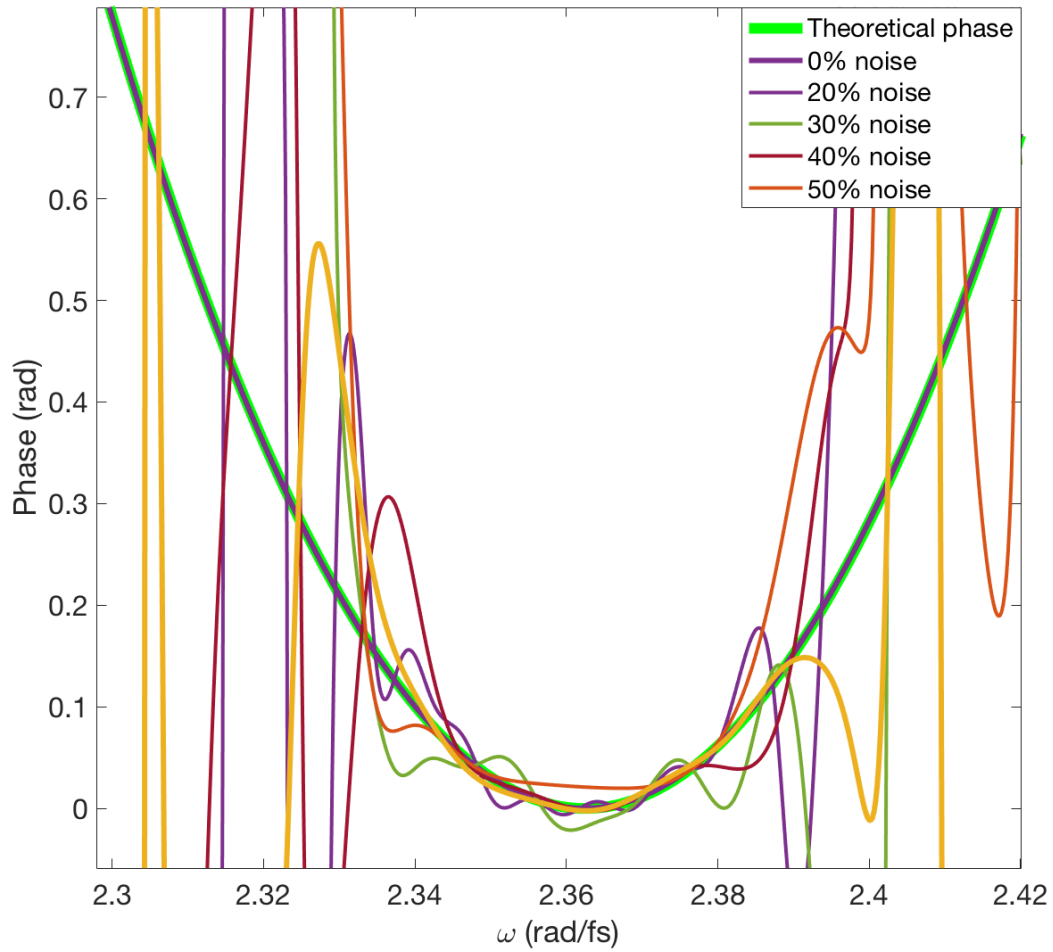


Fig. 5.10 TP phase reconstruction in function of the additive noise level.

applied to the interferogram. The figures show an excellent agreement between the theoretical and reconstructed phases for all the beams.

In order to further analyze the algorithm performance in presence of noise, the behavior for the MICE error as defined in Eq. 5.4 as a function of the additive noise level is given in Fig 5.9, which underlines an overall linear trend.

Moreover in Fig. 5.10 the reconstruction of the TP phase is shown for the same noise levels; the phase is always retrieved but as the noise level increases the

reconstruction is accurate only in the region where the beam intensity is sufficiently high.

5.3.2. FILTERED TP SIMULATION

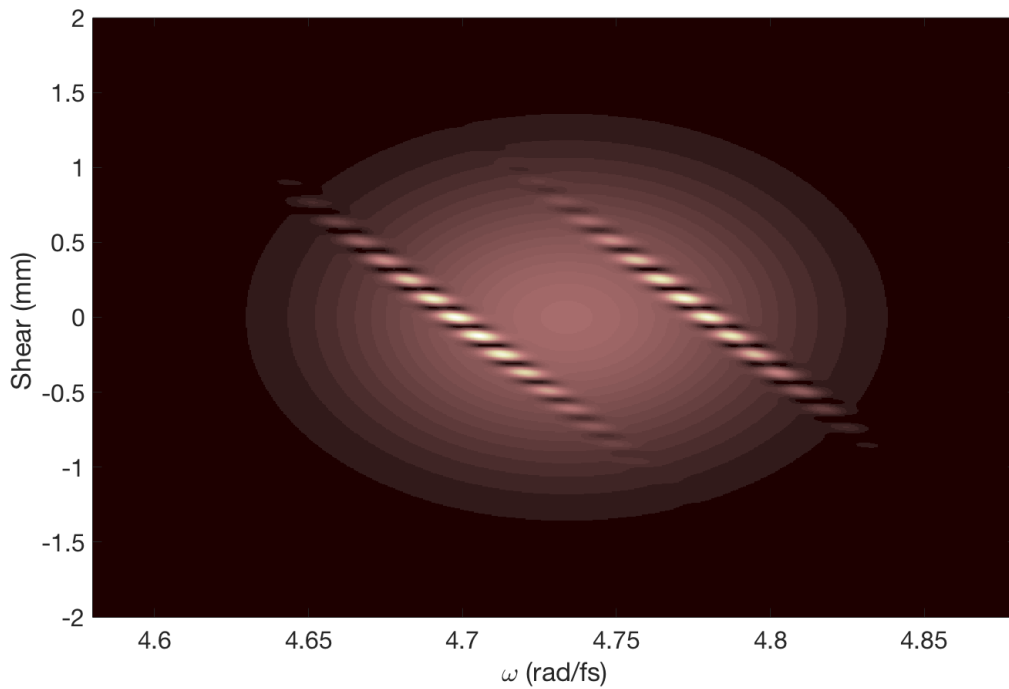


Fig. 5.11 Simulated SPICE interferogram of a filtered TP

The same simulation is repeated but this time the TP spectrum filtered so that the two spectral Gaussian components have a FWHM of $\sigma = 10 \text{ px}$ and the spectral gap is $\simeq 120 \text{ px}$ wide. The interferogram so obtained is shown in Fig. 5.11.

As in the unfiltered case, a 2-dimensional FFT is performed on the interferogram, the sideband isolated and inverse Fourier transformed back. In Fig. 5.12 the interferogram, the simulated AC, and the reconstructed AC are shown for 0% and 50% additive noise level (left and right column). Also in this simulation the sideband,

which bears the characteristic two-lobed structure, is well reconstructed even in the presence of extremely high noise.

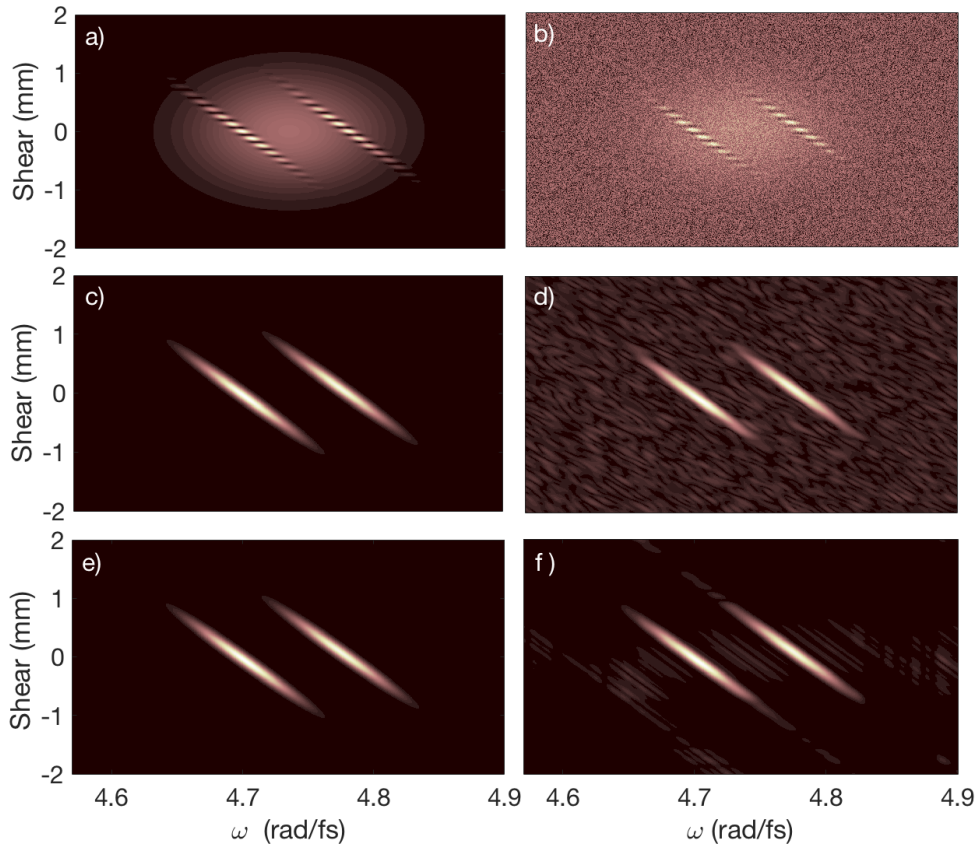


Fig. 5.12 Left column: interferogram (a), AC side band (c), retrieved AC side band (e) in the absence of noise; Right column interferogram (b), AC side band (d), retrieved AC side band (f) with 50% additive noise

The MICE error in Fig. 5.13 shows in fact a trend similar to the previous case – however the values are slightly higher and this is probably due to the fact that the lower amount of points imposed by the TP's structure result in less redundancy than before.

In Fig 5.14 the reconstructed phases for the TP, RP and AP are shown together with the theoretical ones and the agreement between the two is excellent; in

particular we notice how the relative phase between the two components of the TP is correctly reconstructed.

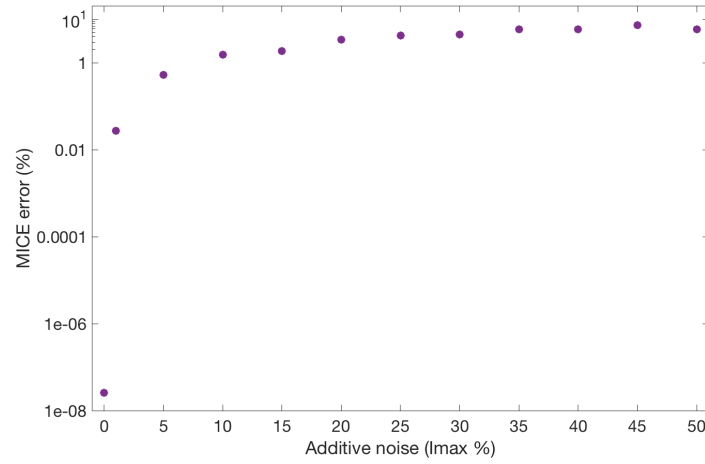


Fig. 5.13 Semi-log plot of the MICE error versus of the additive noise level.

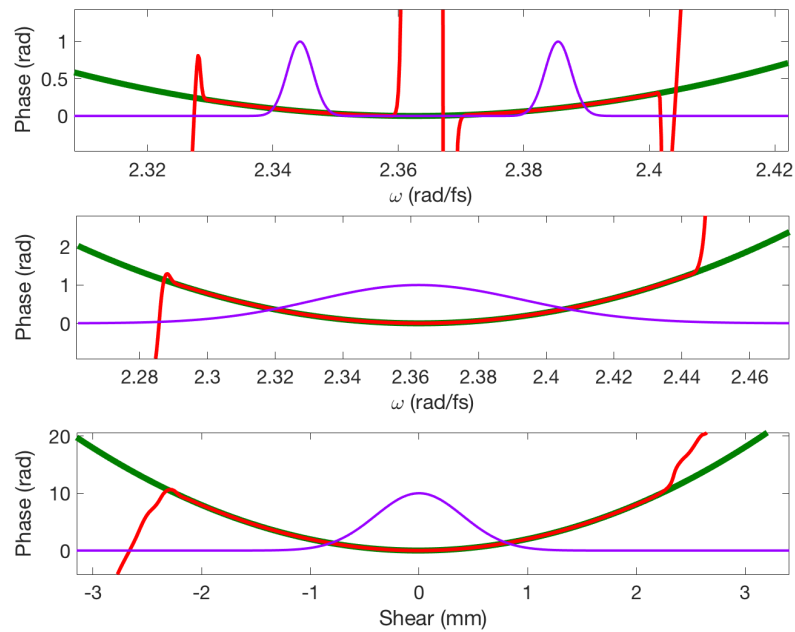


Fig. 5.14 Theoretical (green) phase , reconstructed (red) phase, and spectrum (purple) for the TP (top panel) RP (middle panel) AP (lower panel)

The reconstruction of the TP phase as a function of the additive noise level is shown in Fig. 5.15: despite the noisy structure and the reconstruction only for higher spectral intensities, the relative phase is successfully reconstructed even with high noise levels.

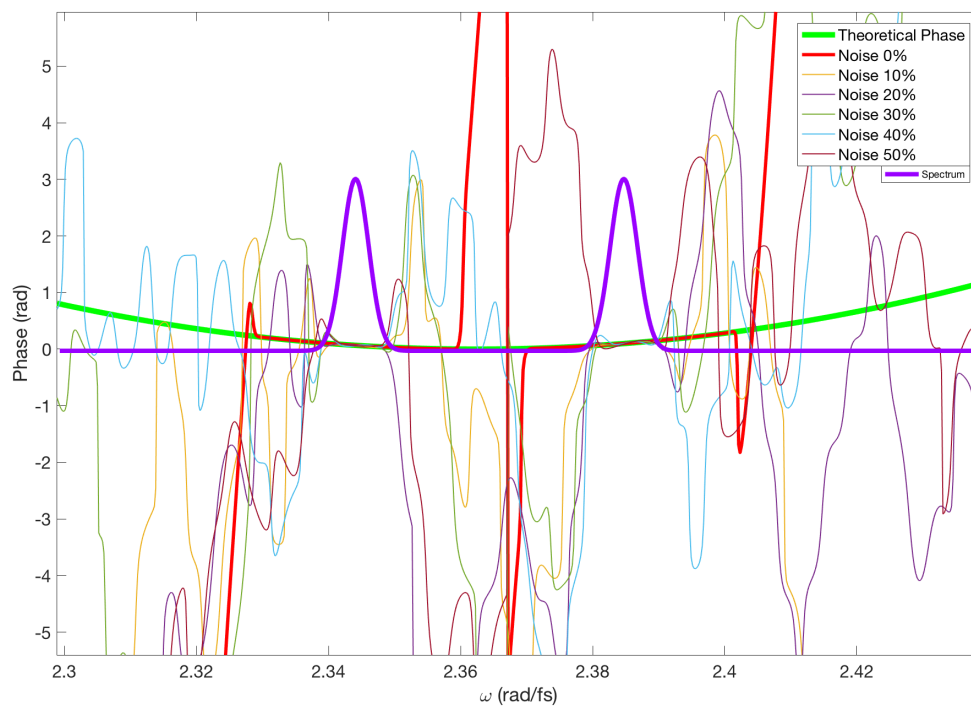


Fig. 5.15 TP phase reconstruction in function of the additive noise level.

5.3.3. SPACE-TIME COUPLING IN SPICE

A common threat to the success of many pulses characterization techniques lies in STC and alas SPICE is no exception. By creating a coupling between the shear/spatial domain and the time/frequency one, the algorithm fails at correctly identifying which phase to attribute to which field. When STC is present, in fact, the phase of the AC band will read:

$$\Phi_{AC}(\omega, \Omega) = \Phi_{RP}(\omega) - \Phi_{TP}(\omega - \Omega) - \Phi_{AP}(\Omega) + \Phi_{STC}(\omega, \Omega) \quad (5.5)$$

The STC phase term, on the other hand, will be of the form:

$$\Phi_{STC}(\omega, \Omega) = \gamma(\omega - \omega_0)(\Omega - \Omega_0). \quad (5.6)$$

where Ω is mapped onto the space domain and one can usually define $\Omega_0 = 0$ so that (5.6) becomes:

$$\Phi_{STC}(\omega, \Omega) = \gamma\omega\Omega - \gamma\omega_0\Omega, \quad (5.7)$$

with one term depending exclusively on Ω and one depending on $\omega\Omega$.

The term in Ω is linear and thus only provides an additional GD on the ancilla. The term in $\omega\Omega$ instead is what effectively prevents the algorithm from successfully retrieving the phases. In fact, the algorithm interprets this term as a function of ω , Ω , and $\omega - \Omega$ which are the directions of the three fields. This coupling term is identified as:

$$\gamma\omega\Omega = \frac{\gamma}{2}\omega^2 + \frac{\gamma}{2}\Omega^2 - \frac{\gamma}{2}(\omega - \Omega)^2, \quad (5.8)$$

hence introducing a second order term in each of the three variables. Since the RP, AP and TP depend respectively on ω , Ω , and $\omega - \Omega$, it is impossible for the algorithm to discriminate between the second order terms here introduced and the actual GDD of each field. Although this provides a limitation for this technique, since the effect of the STC on the phase retrieval is described by (5.8), it can be taken into account if the coefficient γ is known, by simply subtracting $\frac{\gamma}{4}$ from the GDD value obtained in the reconstruction, or can be calibrated prior to undertaking a measurement.

5.3.4. OUTLOOK

I have presented in this chapter the theoretical realisation of a new interferometric technique that allows the retrieval of pulses with spectral nulls. There are numerous application which can benefit from such a technique, as discussed in chapter 3. Furthermore this technique also serves as an example for the MICE algorithm capabilities and versatility. In particular it exploits the benefit of not needing a characterised reference, which can compensate for low power in the TP. The algorithm has proven very robust to noise and shows also a quick convergence. One of the main limitations of this new technique is the behaviour in presence of STC, which needs to be calibrated beforehand to obtain a reliable reconstruction.

THE SPICE EXPERIMENT

"SPICE up your life."

- Viva Forever, Spice Girls

In this chapter I present the experiment that I have realised as a proof of principle demonstration of the SPICE technique. The experiment is realised in a test-bed configuration, where the TP shaping can be switched easily between having a gap or a compact spectrum. This has multiple functions: it allows for the calibration against STC with a commercial device, and allows easy comparison of the phase retrieved in the two different scenarios. This has provided a resourceful benchmark for validating the reconstruction of the gapped TP, as will be discussed in the result section.

6.1. SETUP DESIGN

The experiment has been conducted using a 160 mW pick-off from a Ti:sapphire chirped-pulsed amplifier delivering 30 fs pulses at a 1 kHz repetition-rate with $\omega_0 = 795nm$. The pick-off is divided into three beams (TP, AP, and RP) by a series

of half wave plates (HWP) and polarizing beam splitters (PBS) to adjust the power as needed.

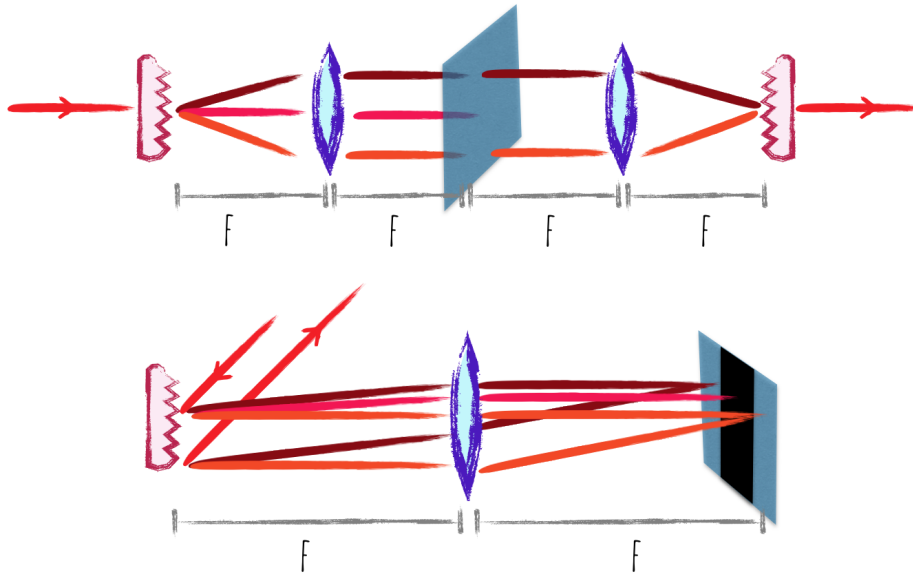


Fig. 6.1 4f-line. Upper panel: scheme of a 4-f line (zero dispersion line) where a mask is put in the fourier plane of the lenses. Lower panel: the same 4-f line in a folded geometry.

The experimental setup has been designed to test the SPICE method, and includes the facility to create TPs with adjustable spectral gaps. This is achieved with a a folded 4f line (4F) geometry as described in Fig. 6.1: by placing a mask in the Fourier plane of the lens, part of the component can be blocked so that only some frequencies recombine in the outgoing pulse. The mask used had triangular structure so that by adjusting its height it was possible to tune the width of the gap and of the pulses. This allowed for the generation of pulses with spectral gaps of different widths and also to easily remove the mask to compare the results with and without a gap present. In the gap example considered, the mask was set to produce a 12.9 nm gap between two peaks each with a 4 nm spectral support; this large gap would cause common reconstruction methods to fail. Importantly, in real-life

scenarios, the TP, AP and RP need not be derived from the same source, since their phases are all estimated by the MICE algorithm without assumptions. This means that it is not required for the power of the TP beam to be high – since it does not need to contribute to the other beams.

This makes SPICE very sensitive to low energy pulses. Of course, the test arrangement used here for convenience does not have this feature, and a TP of 40 μJ energy was used.

The TP is sent through the 4-f line and then focused using a cylindrical mirror (CM1) with $f = 300$ mm into a 100 μm thick type-II BBO crystal (BBO1) (Fig. 6.2). The AP is spatially-chirped using a blazed diffraction grating (G) and is then collimated with a spherical lens (L) with focal length $f = 60$ mm into BBO1 as well. The spherical lens serves to collimate the direction diffracted by the grating and to focus the beam in the other dimension, so that it overlaps spatially with the TP.

This choice of focusing produces the same spot size for the TP and AP at the focus, guaranteeing complete spatial overlap. The two beams arrive on the crystal in a non-collinear geometry, with angles $\theta_{TP} = 10^\circ$ and $\theta_{AP} = 6^\circ$ with the surface normal respectively. This way, the phase-matching function allows for sum-frequency generation across the whole spectrum of the TP with all frequencies of the AP spanning the gap as shown in Fig. 6.3 which represents the phase matching for a Type II BBO crystal with non collinear geometry cut at $\theta = 53^\circ$. The realization of a large phase-matching bandwidth is crucial as it must ensure that the spectral gap is bridged, otherwise the SPICE method would fail. In order to accommodate sufficient spectral components within the transverse profile of the TP the chirp-rate must also be taken into account, which in our case is equal to $\alpha = -50.2 \text{ mrad/fs/mm}$. The arrival times of the TP and AP at the crystal are synchronized by controlling their relative delay by means of a delay line (DL1) on the AP path. The RP is

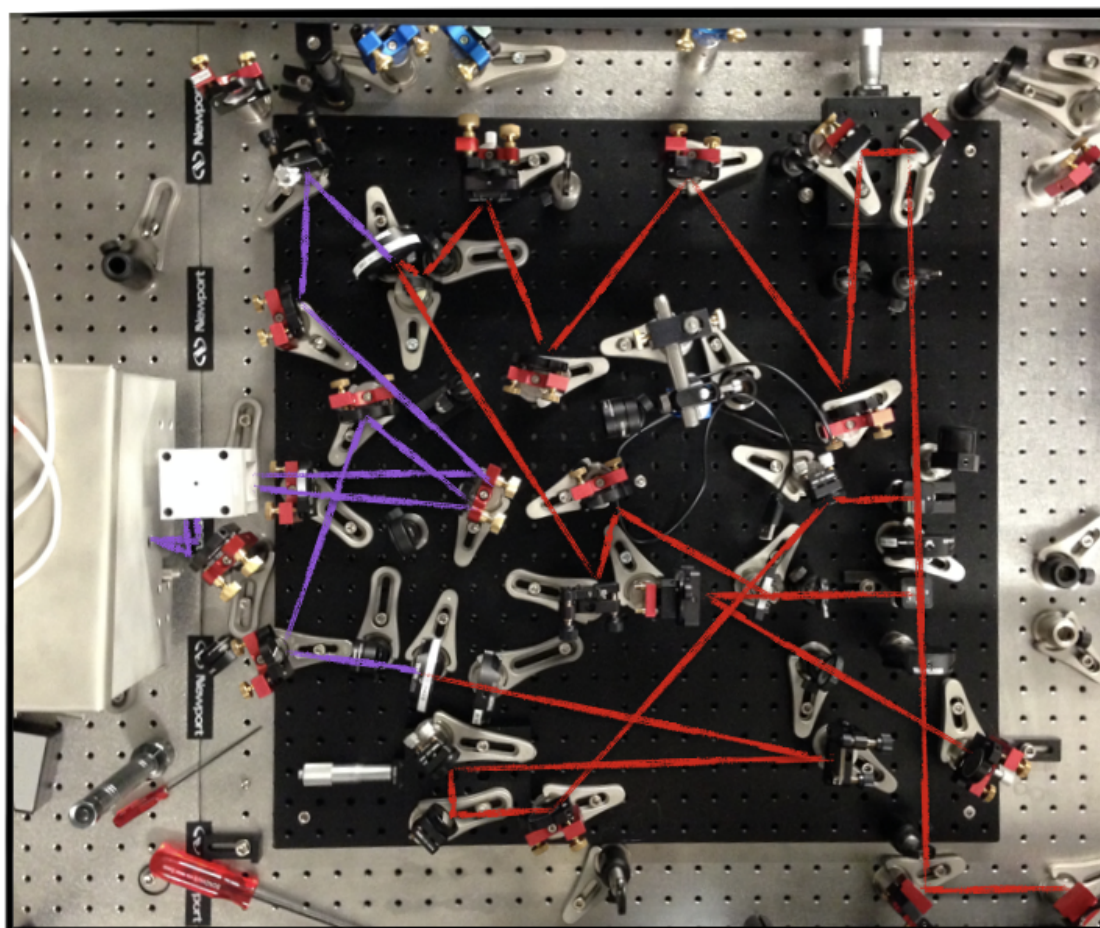
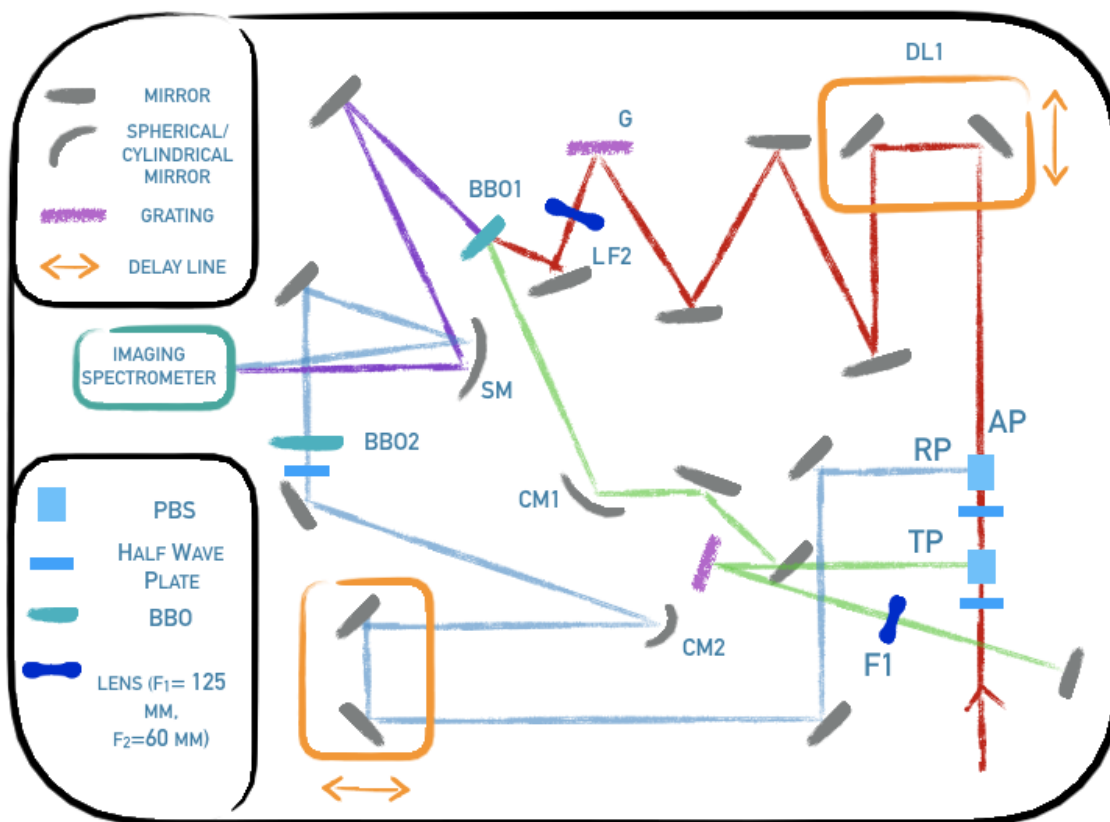


Fig. 6.2 Experimental setup

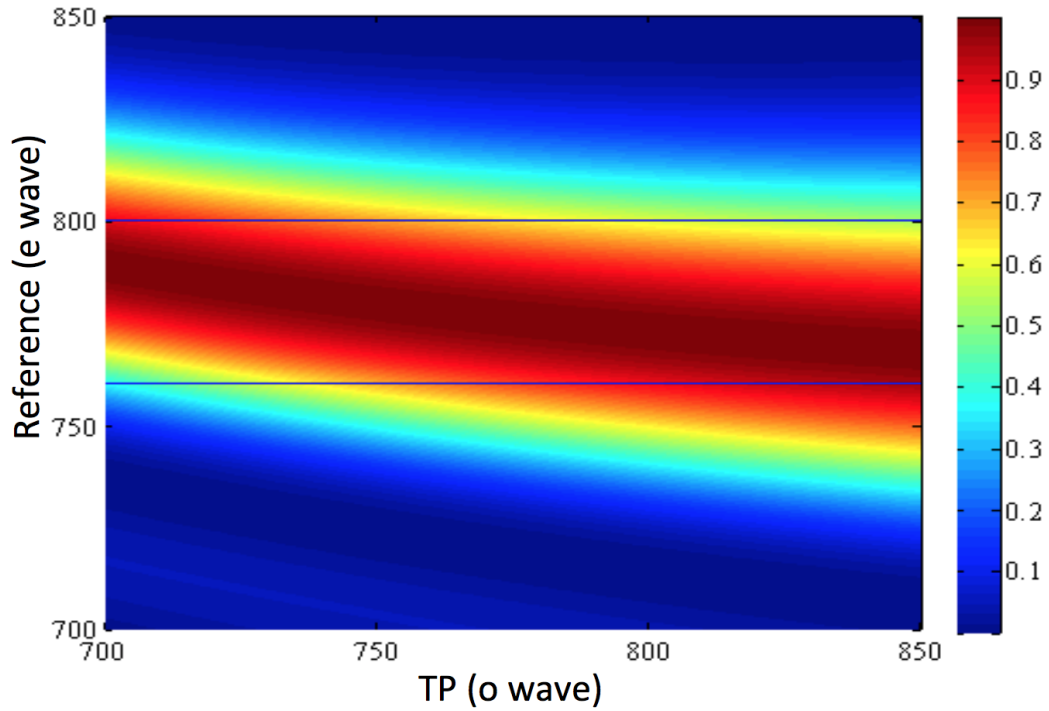


Fig. 6.3 Phasematching of BBO1

focused with a cylindrical mirror (CM2) of $f = 300$ mm into a second $100 \mu\text{m}$ thick BBO crystal (BBO2). In order to upconvert a sufficiently large bandwidth a Type II crystal is used also in this instance; to do so the RP is rotated with a HWP at 22.5° . In Fig. 6.4 the phase-matching function is shown for a crystal cut at $\theta_{\text{BBO2}} = 43^\circ$ for a collinear geometry (the o and e wave are in fact just derived from the H+V polarisation introduced on the RP with a HWP).

The RP and the SP are then spatially interfered on the entrance slit of an imaging spectrometer (IS) using a 2"-diameter, $f=200$ mm spherical mirror (SM). Interferograms are then recoded using a CMOS camera.

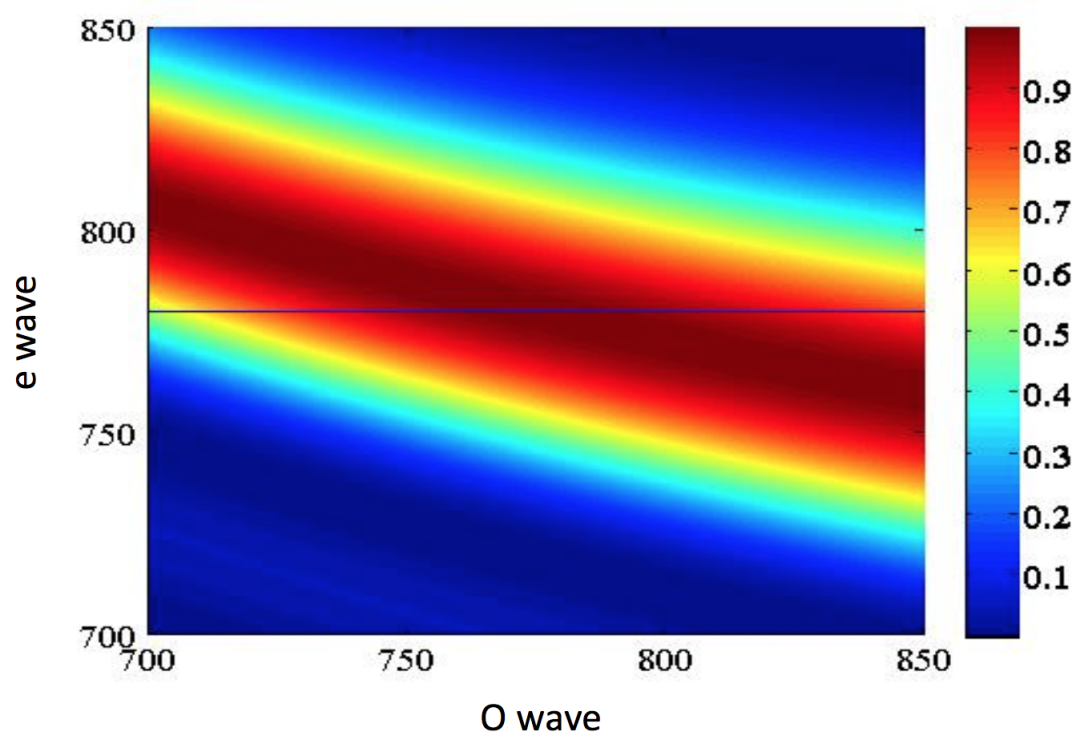


Fig. 6.4 Phasematching on BBO2

6.2. ALIGNMENT AND CALIBRATIONS

- **Spatial overlap in BBO1.** In order to obtain the SP the TP and AP need to be well overlapped in time and space at the SFG crystal BBO1 in Fig. 6.2. For the time component, a stage is used to correct for the beam path length; In order to check the spatial overlap the components are focused with the system described in the previous section ensuring that they arrive on the crystal with a similar spatial profile, and a camera is placed in the crystal position, as shown in Fig. 6.5 a). The AP, having more experimental constraints due to the grating and lens alignment, is kept fixed and the TP is displaced vertically to be overlapped to the AP (Fig. 6.5 b)).

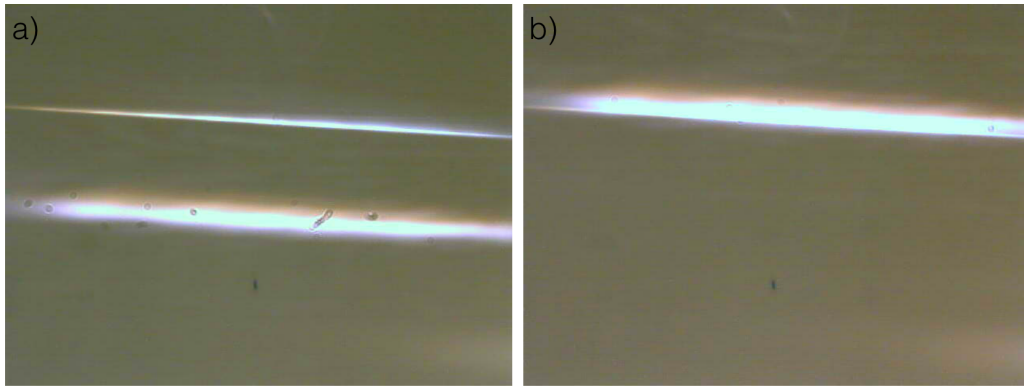


Fig. 6.5 a) Spatial profiles of the AP and TP at the position of the SFG crystal (BBO1 in Fig. 6.2) b) Overlapped spatial profiles of AP and TP

- **Delay lines.** In order to obtain the SP pulse the TP and AP path difference is adjusted by a delay line assembled in the AP path. For the SP to interfere with the RP there is need to match the path length also between the SP and the RP, which is done, once the upconversion of the SP is correctly implemented, via a delay line (DLR in Fig 6.2) on the RP path. This was a critical alignment procedure as the setup geometry has several mode matching constraints due

to phase matching geometries and to the spatial interference, and required considerable experimental effort to accomodate all the critical parameters.

- **Spectra acquisition.** MICE presents an ambiguity due to the pulse being retrieved up to an exponential factor: $e^{(\beta_1 + i\beta_2)\omega}$, with $\beta = \beta_1 + i\beta_2$, $\beta_1, \beta_2 \in \mathbb{R}$ (see Chapter 3), which results in an ambiguity on the absolute phase of each pulse due to the complex term – as with other species of SPIDER – and, more crucially, on their amplitude due to the real term. While the absolute phase has no importance for pulses which are not CEP-stabilized, it is desirable for a reconstruction method to automatically retrieve also the spectra of the pulses. This can be achieved in SPICE by recording separately the spatially-resolved spectra of the individual arms that arrive on the imaging spectrometer to form the interferogram, that is SP and RP pulses. The captured images for the spectra are shown in the case of the unfiltered SP and RP in Fig. 6.6 and filtered SP and RP in Fig. 6.7.

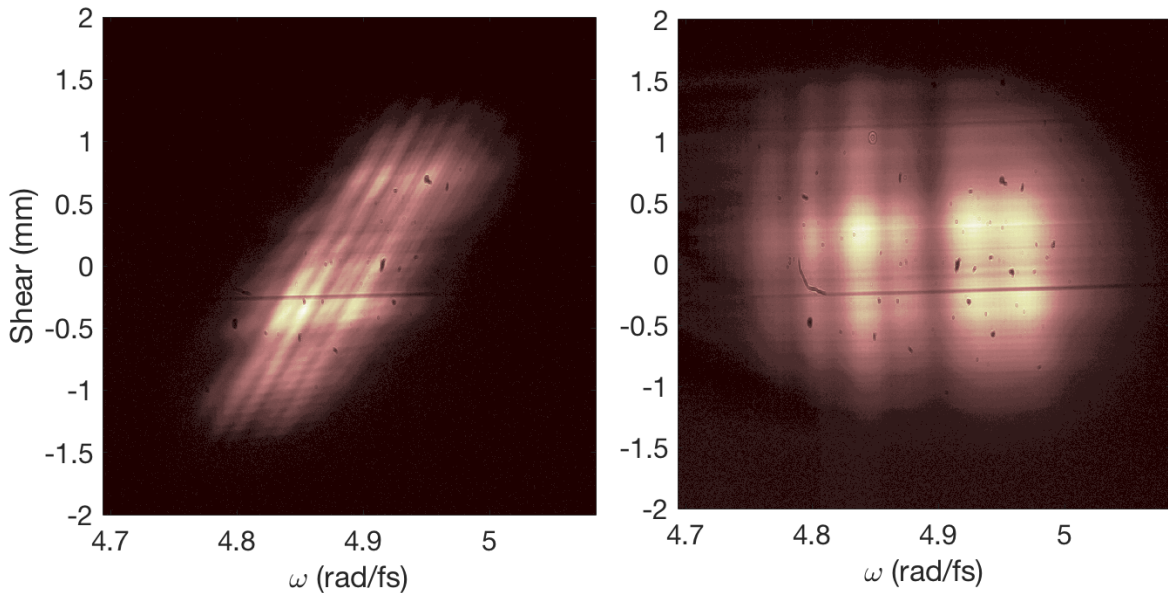


Fig. 6.6 Spatially resolved SP (left) and RP (right) spectra for the unfiltered TP measurement

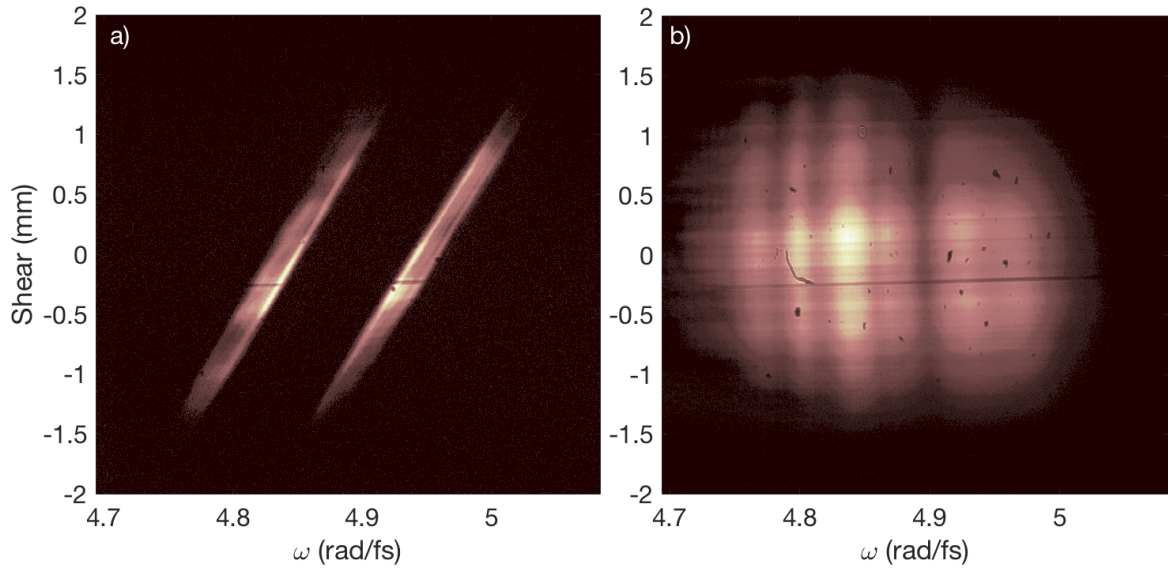


Fig. 6.7 Spatially resolved SP (left) and RP (right) spectra for the filtered TP measurement

- **shear calibration** As with methods like SEA-CAR-SPIDER, there is a need to perform a shear calibration on the SP prior to reconstructing any phase information from the interferogram.

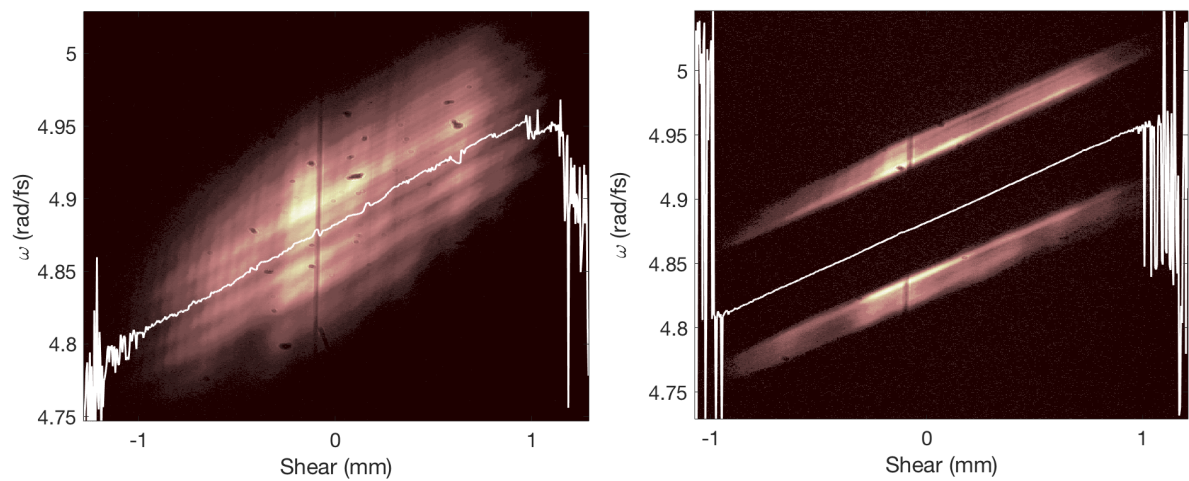


Fig. 6.8 Shear calibration of the SP for the unfiltered TP (left panel) and filtered TP (right panel).

Differently from the SEA-CAR SPIDER this is necessary only for one arm, that is the SP arm, as the RP does not undergo any shearing process. This can be done by utilizing the same spatially-resolved SP spectra which is in any case acquired for the spectra retrieval. The shear calibration is performed by measuring how the SP central frequency varies with the position of the beam. The chirp rate for our setup, calculated from the fit on the shear calibration, is $\alpha = -50.2 \text{ mrad/fs/mm}$. The results of the calibration are shown in Fig. 6.8 both for the unfiltered and filtered TP case.

- **STC calibration:** As was discussed in the previous chapter the presence of STC introduces an additional quadratic phase in both the RP and SP arm. Unfortunately the arrangement chosen for the setup is also a source of STC due to the non-collinear asymmetric geometries. This means that even in the case of an STC-free beam, the setup needs calibration of STC to be reliable. To do so we perform with the SPICE setup the measurement of the unfiltered spectrum, and then the same measurement with a commercial SPIDER device. The phase difference between the two methods is to be attributed to the setup STC and is used as a calibration for the phase of the filtered TP. For convenience in the explanation of the extraction procedure, the two measurements will be treated and presented in parallel in the next section, as the procedure in the extraction up until the calibration step at the end is exactly the same.

6.3. RESULTS

6.3.1. SPECTRA RETRIEVAL

For the pulses described in section 6.1, we obtained 2D images of the individual pulses on the imaging spectrometer. They allow to retrieve the spectra of the TP, RP and AP beams in both the filtered and unfiltered scenario.

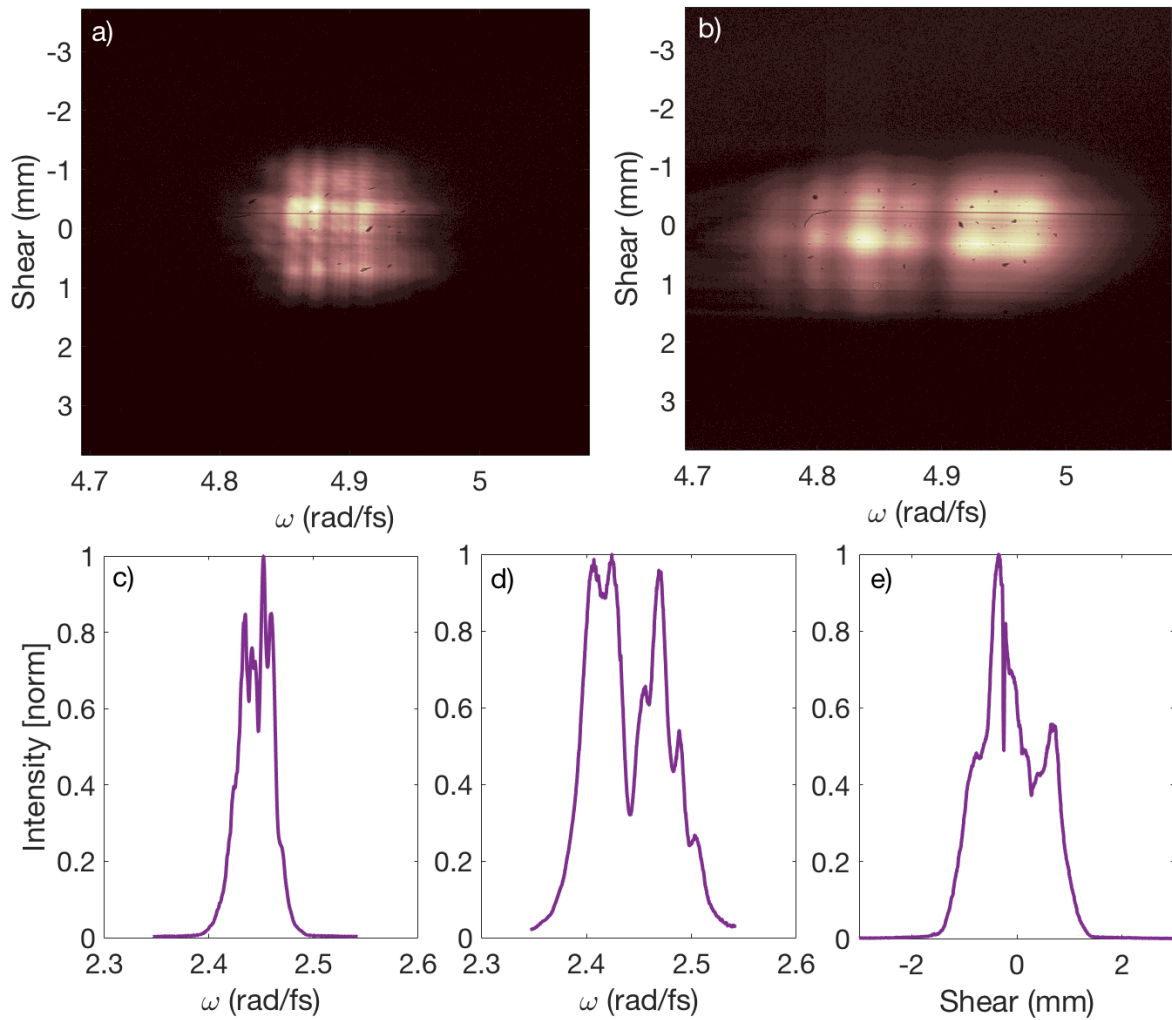


Fig. 6.9 Spectra retrieval: unfiltered TP. a) SP arm after shear is removed b) RP arm c) Retrieved TP spectrum d) Retrieved RP spectrum e) Retrieved AP spectrum

In fact, once the shear is calibrated it is possible to operate a row-rotation on such images obtaining the ones in Fig. 6.9 and 6.10 panel a) and b) to obtain a "shear-free" image, and then by an iterative procedure the spatial and spectral components are retrieved, obtaining the three beams spectra, plus the spatial component of the RP beam which is not of any interest for the reconstruction and thus is not reported in Fig. 6.9 and 6.10.

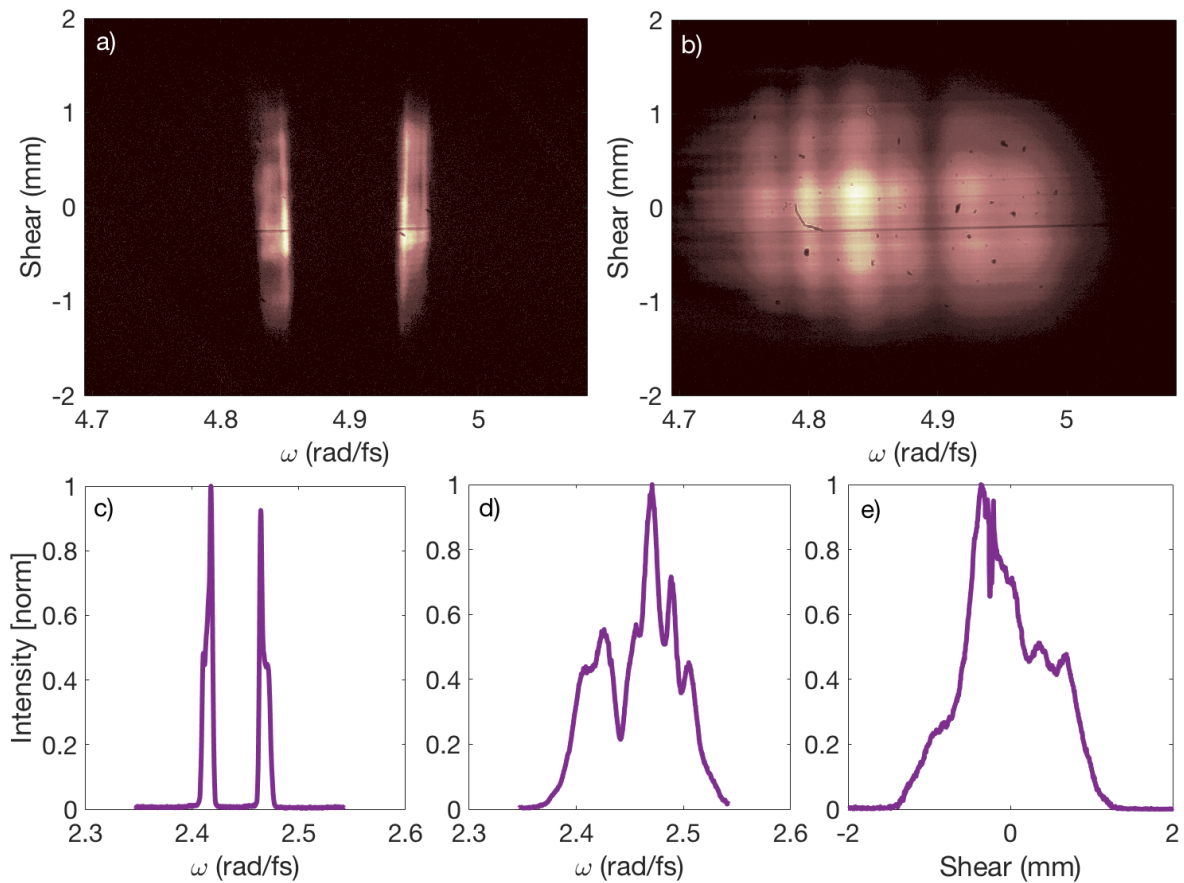


Fig. 6.10 Spectra retrieval: filtered TP. a) SP arm after shear is removed b) RP arm c) Retrieved TP spectrum d) Retrieved RP spectrum e) Retrieved AP spectrum

6.3.2. PHASE RETRIEVAL

The interferogram for the unfiltered and filtered TP, measured with the imaging spectrometer and averaged over 80 ms (i.e. 80 pulses) are shown in Fig. 6.11 and Fig.6.12 a).

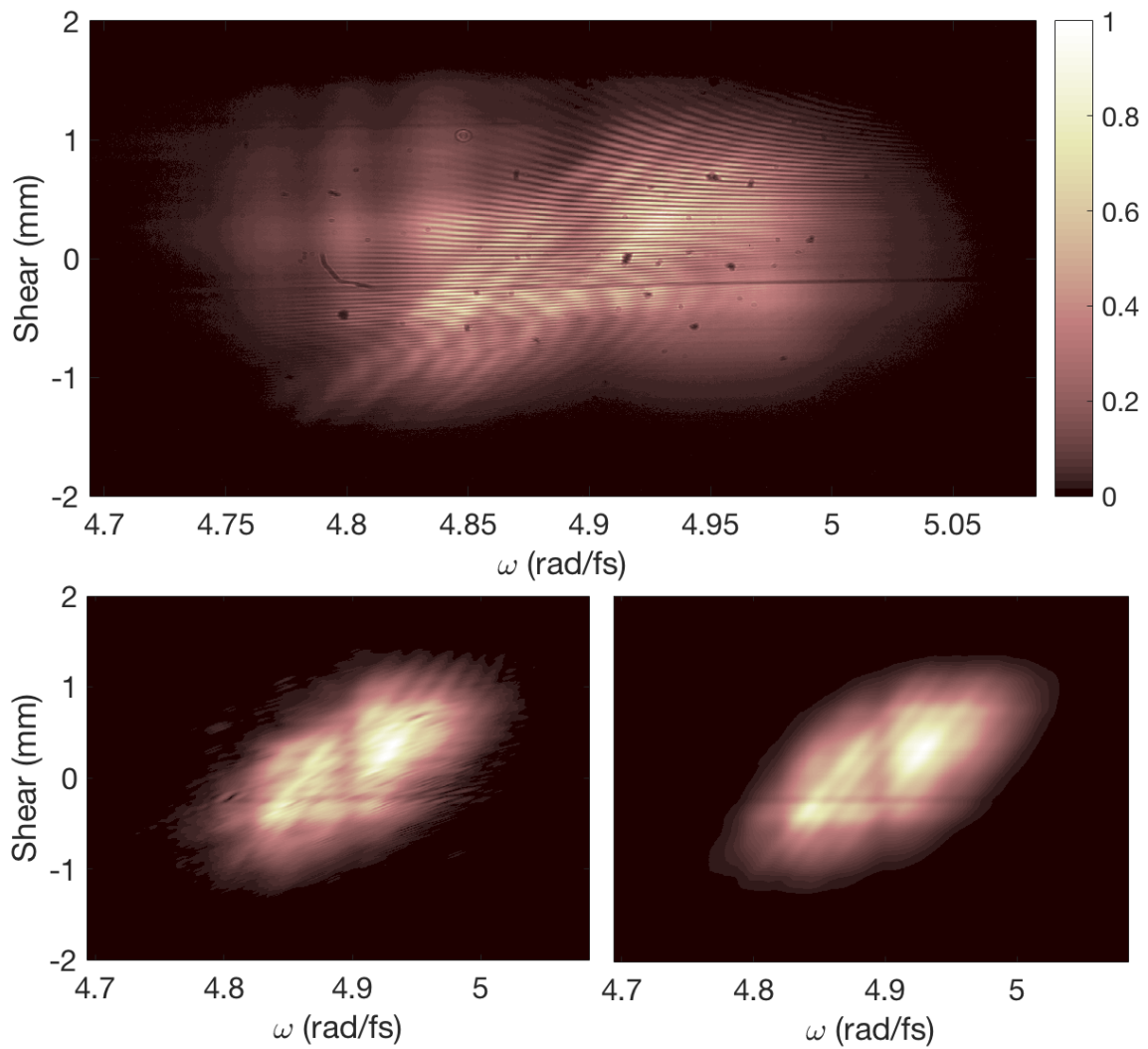


Fig. 6.11 Top panel: SPICE interferogram; Lower left panel: Extracted AC component; Lower right panel: Retrieved AC component.

In the panels b) and c) of Fig. 6.11 and Fig. 6.12 instead the AC components measured and reconstructed are shown. In order to isolate the AC components, the

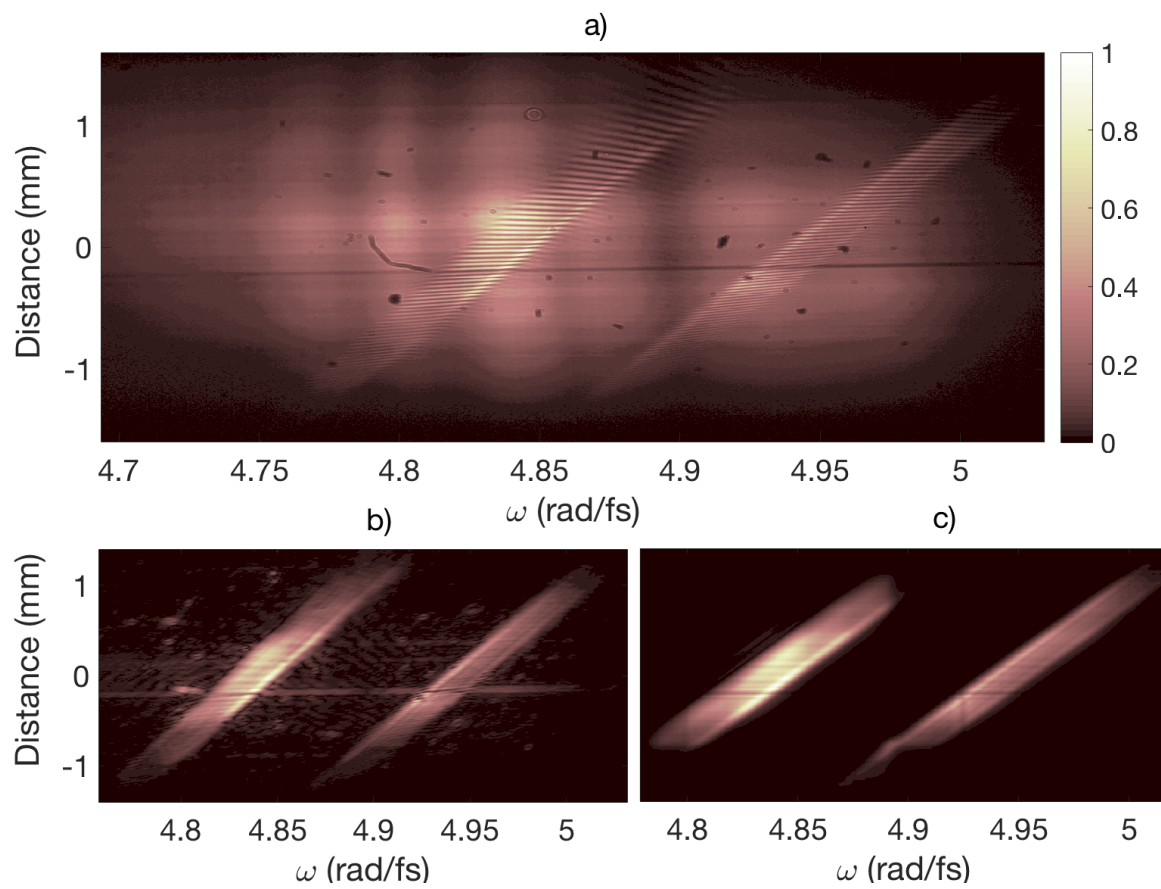


Fig. 6.12 Top panel: SPICE interferogram; Lower left panel: Extracted AC component; Lower right panel: Retrieved AC component.

2D Fourier transform is performed on the interferogram (Fig 6.13) and the sideband is selected by means of an adjustable mask (as represented by the purple rectangle).

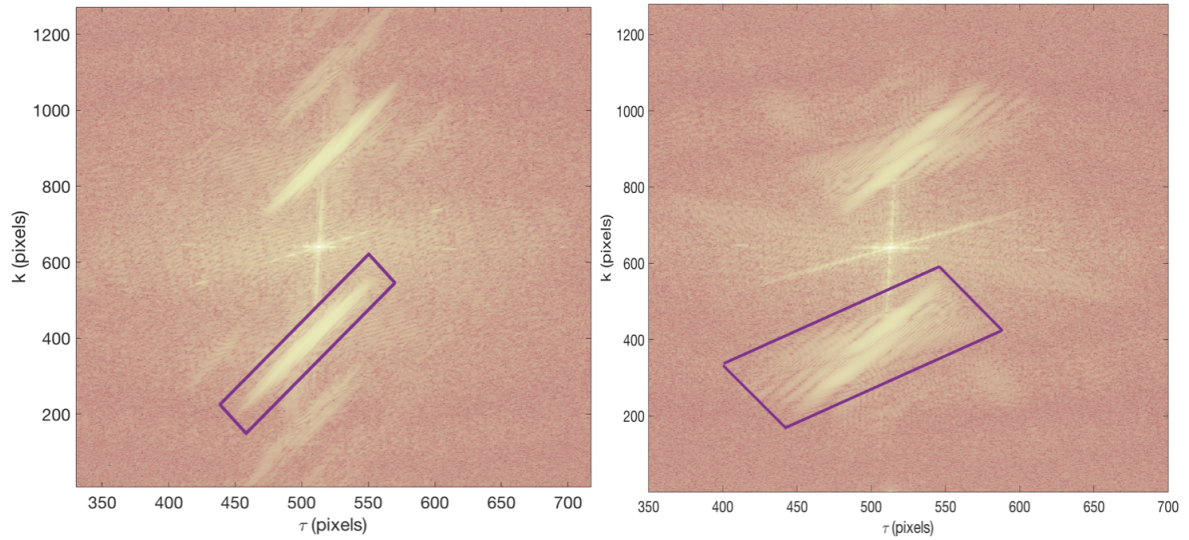


Fig. 6.13 Left panel: 2D FFT of the interferogram for the unfiltered TP. Right Panel: 2D FFT of the interferogram for the filtered TP. The sidebands are highlighted by the purple rectangles

Using the Eq. 5.3 in Chapter 5 on the AC component shown, the MICE algorithm is performed. Both with and without the gap the agreement between the measured and reconstructed AC is excellent and the reconstruction was achieved after just 10 iterations with a MICE error of 0.18% and 0.22% respectively for the unfiltered and filtered case, demonstrating the rapid convergence of the MICE algorithm and the high quality of the reconstruction.

In order to obtain the phase we have first to perform the calibration against STC. To do so, we use the phase difference between the measured phase of the unfiltered TP and the same phase measured with a commercial SPIDER, to calibrate the RP and AP of the unfiltered pulse as well as all the phases of the filtered one. The spectral phases and amplitudes obtained for the TP, RP and AP are reported in Fig. 7.7.

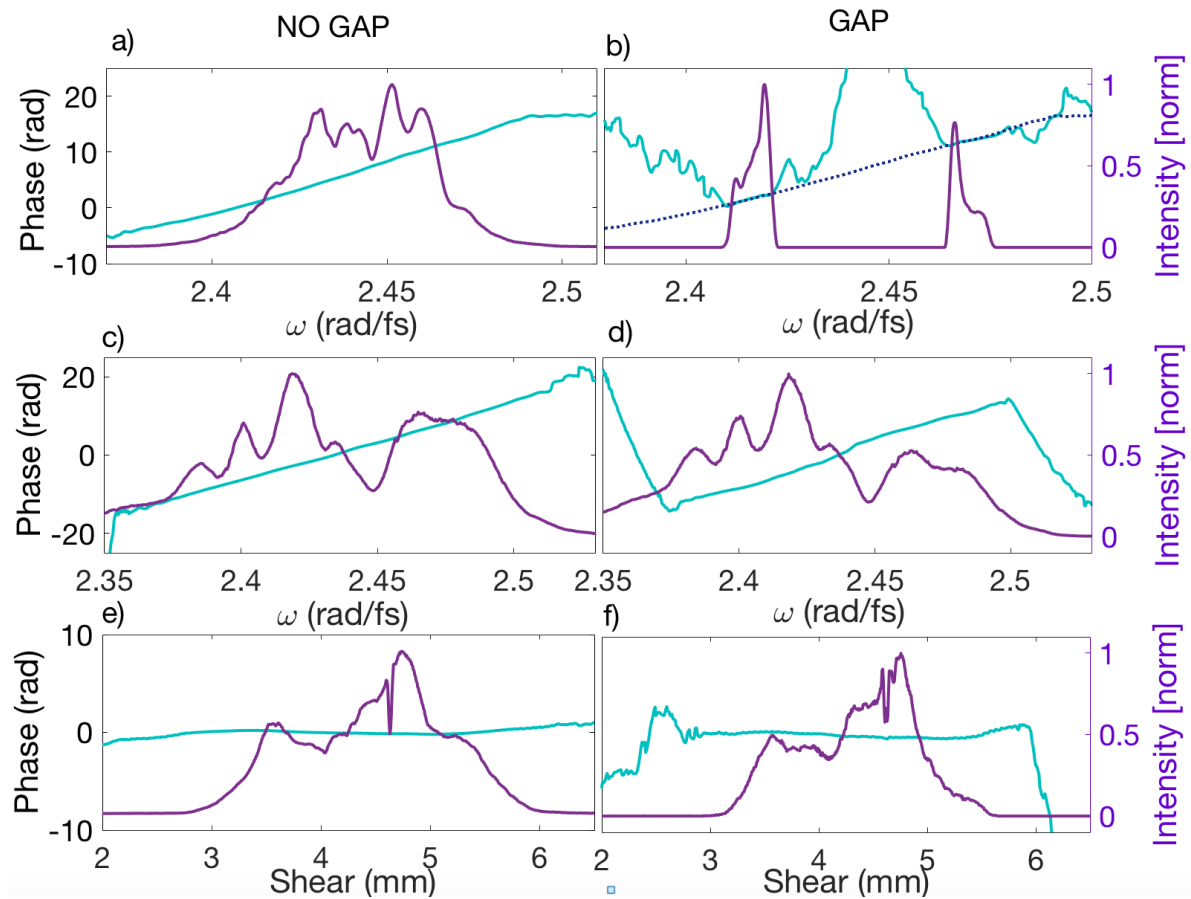


Fig. 6.14 The reconstructed spectral phase function (blue) and amplitudes (red) with and without the gap present. (a-b) TP (c-d) RP (e-f) AP. The key result is highlighted in (b), where the reconstructed phase for the filtered TP is overlapped to that in (a) (in cyan), showing an excellent agreement. The phase in (c-d) is only reconstructed where there is interference with the SP, which is narrower in (d), resulting in the reconstruction of a smaller portion of the RP phase.

The key result is shown in Fig. 6.14 b), where the reconstructed spectral phase (solid blue) function in the regions of the two spectral peaks is in close agreement with that of the unfiltered TP (dashed cyan). The SPICE technique is thus shown capable of reconstructing the relative phase of two well-separated spectral components, which positively demonstrates the objective of this thesis, namely "bridging the gap". There are two features worth noticing in the reconstruction: the first is that the relative phase between the two peaks of the TP is reconstructed up to a multiple of 2π , which have been removed in the graph to compare it with the unfiltered phase. This gives no physical effect. The second is that interference does not occur along the whole spectrum of the RP: this means that the phase reconstruction of the reference will only be consistent within the bandwidth where fringes are present; since this is the only relevant part of the spectrum, this does not hinder the reconstruction of the TP, which is our ultimate goal.

6.3.3. CONSISTENCY CHECKS

Although it is evident from the result presented that the relative phase is accurately reconstructed in the presence of a large gap, in order to validate the method we must check the consistency of the phase obtained with the filtered pulse against that of the unfiltered pulse, since there should be no difference between the two. As the TP spectrum is quite narrow, it is not very reliable to perform a fit on the phase to check the consistency of the GDD, as there aren't enough point for the fit to produce an accurate and reproducible result. For this reason it is convenient to switch to the temporal domain where the effect of the phase is evident on the whole structure of the pulse. For the filtered TP the temporal profile is obtained by Fourier transforming the pulse: $P_1 = S_{gap} \cdot e^{i\Phi_{gap}}$, where S_{gap} is the spectral amplitude of the filtered TP, and Φ_{gap} is its phase.

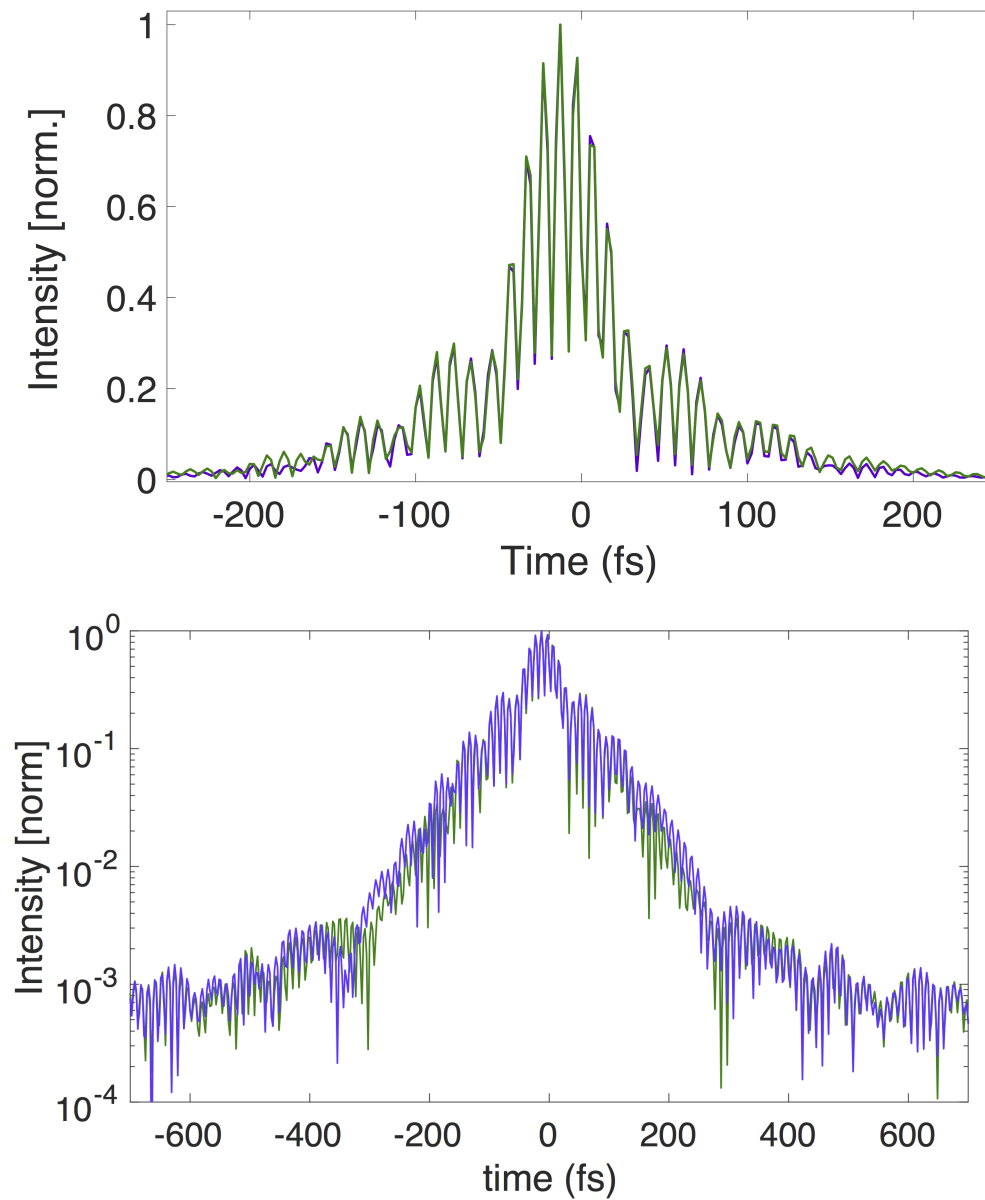


Fig. 6.15 Comparison between the temporal profiles of the pulses P_1 (violet) and P_2 (green) in linear (top panel) and logarithmic (lower panel) scale, obtained using the same spectral amplitude of Fig. 7.7 (b) and the phases shown in Fig. 7.7 (a) and (b) respectively.

However in order to have a meaningful comparison between the phases reconstructed this cannot be directly compared to the temporal pulse of the unfiltered TP as the two spectra are different and so the temporal profiles would differ regardless of the phases. The only possible fair comparison is to use a fictitious pulse which has the same filtered spectrum of P_1 , but bears the phase obtained with the unfiltered reconstruction, that is the Fourier Transform of $P_2 = S_{gap} \cdot e^{i\Phi_{no-gap}}$, where Φ_{no-gap} is the phase of the unfiltered TP. These temporal profiles show an overall duration dictated by the full width of the spectrum, modulated by a fast oscillation whose period is linked to the presence of the spectral gap as well as to the chirp. We can observe in Fig. 6.15 a good agreement between the two temporal profiles wherever the intensity is a substantial fraction of the peak value. This is reflected in a root mean square error (RMSE) of 0.57%, demonstrating how the relative phase between the two peaks has been correctly reconstructed.

A further consistency check carried out was the measurement of the GDD of a glass window inserted at the entrance of the interferometer. The window used was a 5mm fused silica flat with $GVD=36.16 \text{ fs}^2/\text{mm} \cdot \text{rad}$ at $\lambda = 785 \text{ nm}$, resulting in a group delay dispersion of $GDD=180.8 \text{ fs}^2/\text{rad}$. In Fig. 6.16 the phase difference between the TP phases measured with and without glass window is shown, together with the TP spectrum and the resulting fit to the phase difference weighted with the spectrum. The GDD obtained from such fit is $GDD = 180.78 \text{ fs}^2/\text{rad}$, in agreement with the expected value.

6.4. OUTLOOK

In this chapter I have presented the experimental results for the proof-of-principle experiment of SPICE. I have shown that this new technique can successfully retrieve

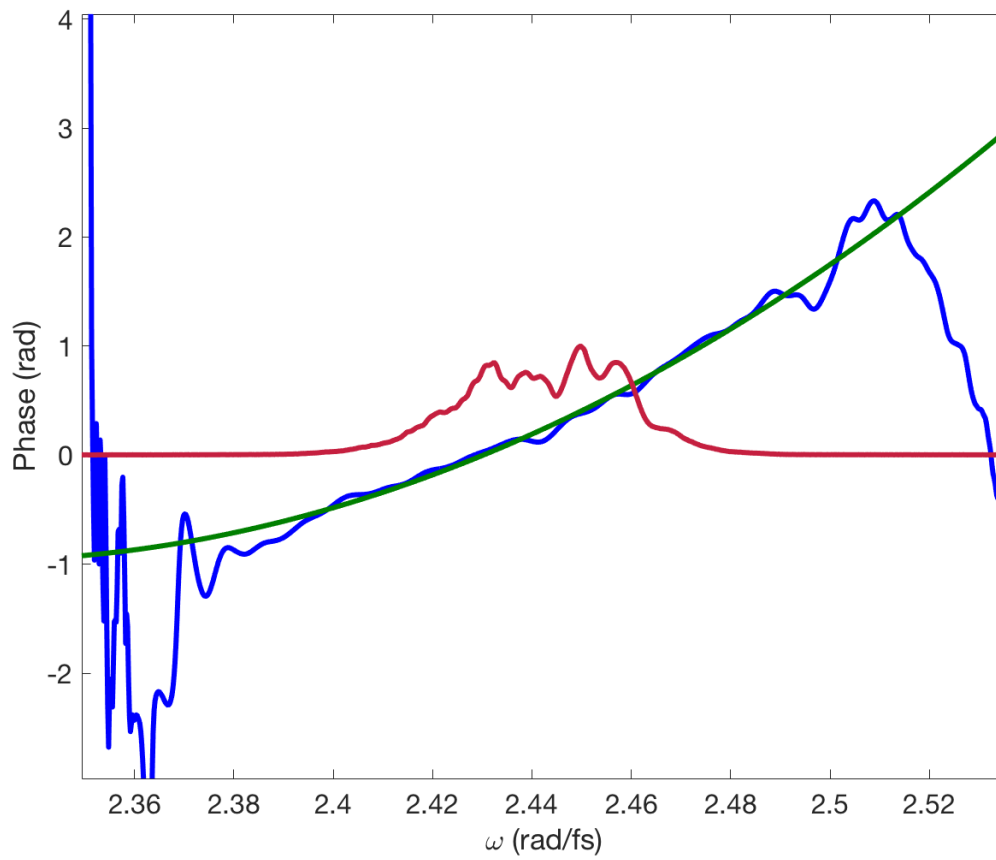


Fig. 6.16 Phase difference between $\text{PHI1}_{\text{glass}}$ and $\text{PHI1}_{\text{noglass}}$.

the relative phase between two separated spectral components. The experimental arrangement used however presents a difficulty due to the introduction of STC, which cannot be properly handled by the algorithm as explained in the previous Chapter. Nonetheless, after performing the required calibration of the STC, the results show that the relative phase is reconstructed and the consistency checks validate the method. Further developement should include finding a STC-free geometry and extending the experimental apparatus to cover multiple gaps.

XUV SPICE

Extensions of MICE to different wavelengths and to spatial characterization are also a possibility and have been carried out in [107], paving the way for a full all-optical spatio-temporal characterization of High-Harmonics Generation (HHG) sources [25, 108].

To have a full characterization however, together with the spatial one performed by Mang et al. in [107] a way to characterize the time-dependent field is required. To this extent Attosecond Resolved Interferometric Electric-Field Sampling (ARIES) [109] has provided an all-optical technique for directly measuring the electric field, $E(t)$, by mapping the HHG cutoff position as a function of the delay between a test pulse to be measured and a more intense probe pulse. For the characterisation in the spectral domain instead, some work on this has been carried out by Mairesse et al. [110], which have tried to implement a version of SPIDER for HHG.

In their work, Mairesse et al. overcome the difficulty of having two identical sheared and time-delayed replicas of the harmonics test pulse needed for the SPIDER technique by shaping the parent infrared pulse with the use of an acousto optical modulator (DAZZLER). The procedure used is pictured in Fig.7.1.

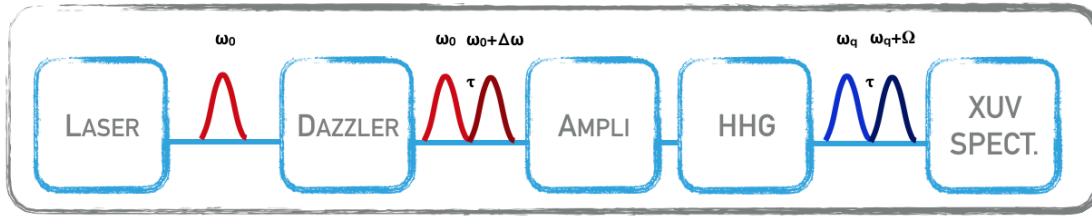


Fig. 7.1 Mairesse XUV SPIDER scheme

The beam with central frequency ω_0 is sent into the DAZZLER, which is programmed to deliver two time-delayed copies. The Dazzler however cannot effectively spectrally shear the pulse, but, if the incoming pulse is sufficiently flat, it can cut two slices of the incoming pulse at an arbitrary distance $\delta\omega$ to look quite the same. As the DAZZLER cannot support too strong of a power, the two copies are then sent to an amplifier system to obtain the power needed for the HHG conversion. The HHG beams created are then measured with an XUV spectrometer.

The main limitations of Mairesse's work are the following:

- 1. The copies utilised are not actual spectrally sheared copies
- 2. The technique allows measurement of single harmonics but fails in the reconstruction of the temporal profile of the whole pulse as the harmonics spectrum is characterized by large intensities gaps.

A MICE/SPICE approach is perhaps the most direct means to solve those two aspects: first of all, being able to characterise multiple pulses at the same time can be used to at least relax the requirement of having two same TP copies to perform the SPIDER measurement; furthermore, if a pulse spectrally-wide enough to span between two harmonics were to be available, an extension of SPICE would allow for it to be used as an unknown reference for the characterisation of the gapped harmonics pulse.

In order to verify if this could be a way forward to a more robust spectral characterization of HHG, I have performed a preliminary experiment which consisted in a MICE-modified version of Mairesse's setup, to measure the infrared parent pulses, as described by the setup of Fig. 7.2.

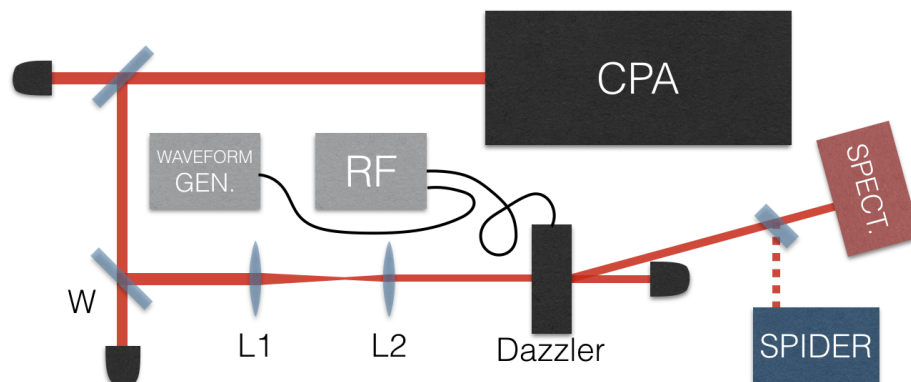


Fig. 7.2 Experimental setup

A 2.2 mW pick off from the CPA is selected and a telescope is set to meet the dazzler entrance requirement. The beam shape after the telescope is shown in Fig. 7.3.

The beam is aligned at a long distance so that it enters perpendicular to the dazzler aperture. The power after the dazzler is $670 \mu W$ for the diffracted beam and $1.3 mW$ for the direct one. A waveform generator triggered by the laser is used to clock the DAZZLER to the laser. The dazzler allows shaping of the incoming beam by applying a modulation both to its amplitude and phase; it also allows for the generation of subsequent pulses (which can be differently shaped) delayed by a chosen amount.

An example of the dazzler program is shown in Fig. 7.4: the orange panel on the left allows for the setting of the parameters amplitude of the transfer function of the dazzler filter. The transfer function has a hyper-Gaussian module of which the

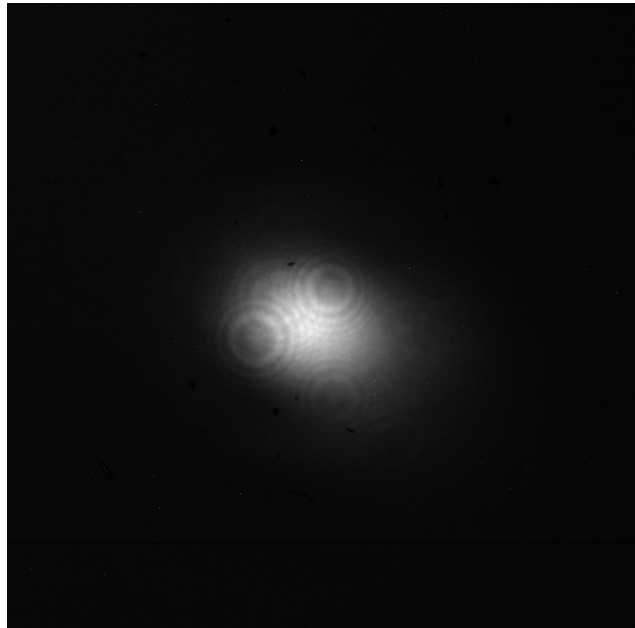


Fig. 7.3 Beam shape after the telescope

central wavelength and width can be decided; furthermore it is possible to produce spectral holes of which position, width and depth can be adjusted.

The green panel allows for controlling of the phase, and is used both to calibrate for the phase added by the dazzler, and for setting an arbitrary phase on the beam, up to the fourth order. The gray panel on the left can be used to add multiple waveforms, which relative delay can be controlled by adjusting the first order phase in the green panel. The two graphs show the resulting waveforms spectrum and time profile. The efficiency of the diffraction is optimized by adjusting the dazzler position so that the beam would fall in the centre of the diffraction region. This has been checked by monitoring the diffracted signal with a photodiode with an input signal programmed so that it would cover the whole delay range.

A frequency calibration was also performed by programming the narrowest possible gap at a given frequency on the dazzler panel and checking the diffracted spectrum with a spectrometer. At the output of the dazzler a flip mirror was

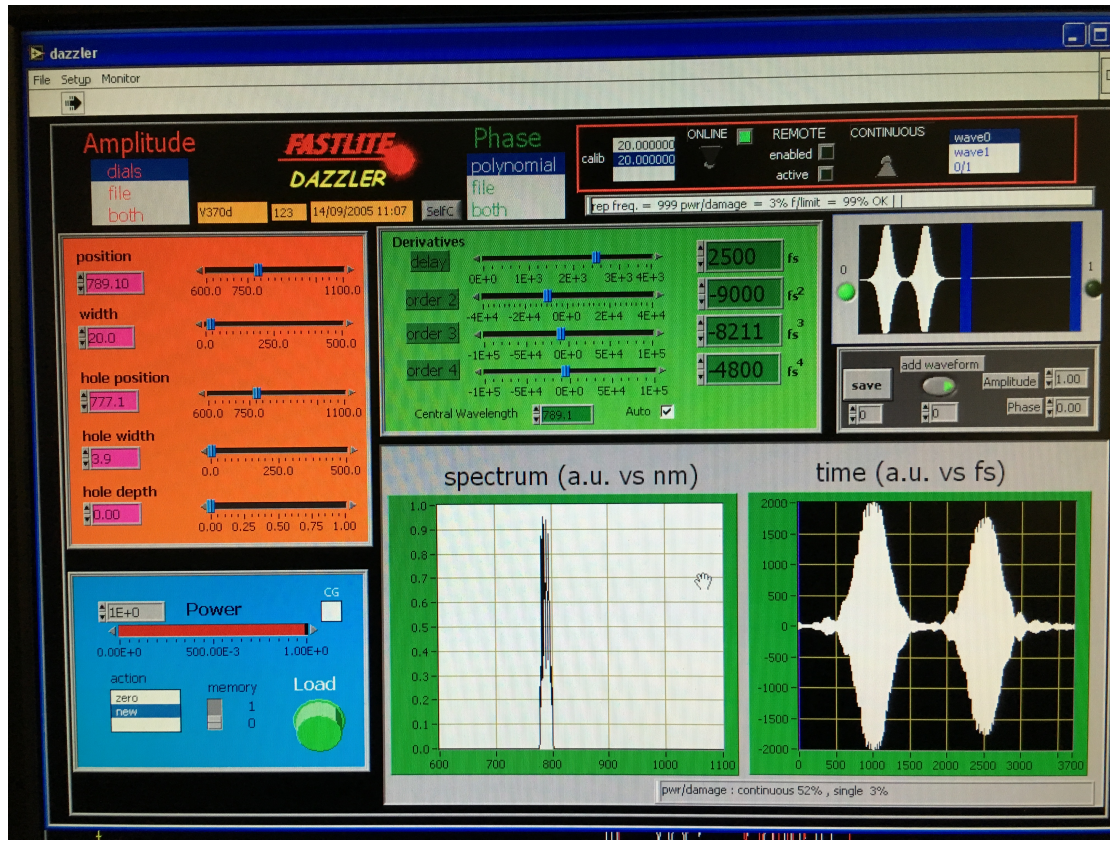


Fig. 7.5 DAZZLER software programmed for MICE experiment.

For each shear an interferogram is recorded using a Thorlabs spectrometer. In Fig. 7.6 a) the combined interferogram of all the subsequent measurements is shown. Much like a 2D measurement, the shear is reported on the Y-axis so that each slice is given by one of the 15 measurements, with the shears going from 0 nm to 3 nm. Fig. 7.6 b) shows the 2D-Fourier transform of the multi-shot interferogram. As seen in the previous chapter isolating the AC band is the first step required to perform a MICE routine.

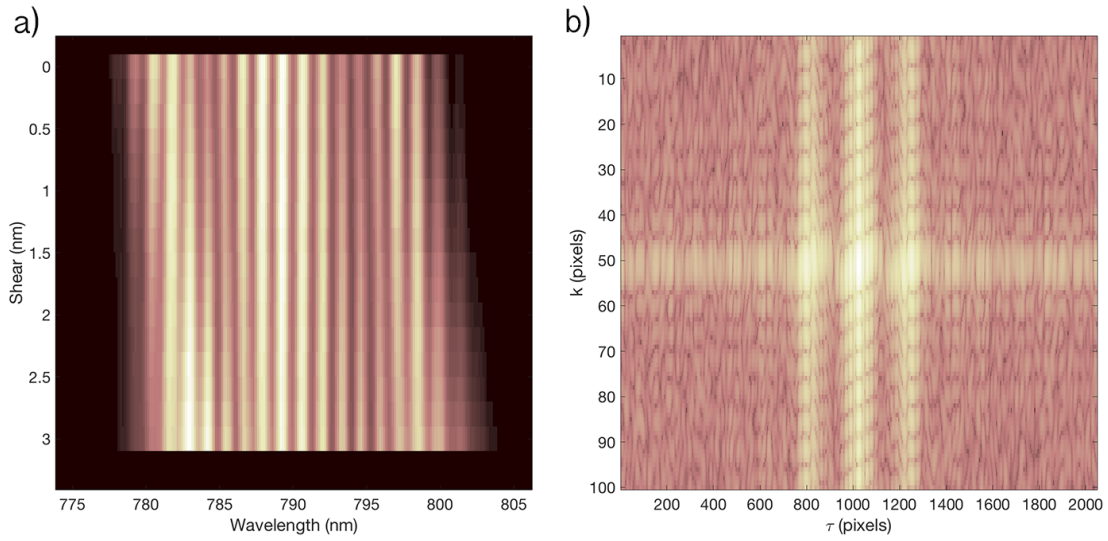


Fig. 7.6 a) Multishot 2D interferogram obtained by varying the shear set with the DAZZLER. b) 2D Fourier transform of the interferogram.

However, contrary to SPICE, in this arrangement there are only two fields involved, namely the two waveforms generated with the DAZZLER – one fixed, and one sheared. Thus, the MICE equation in this scenario becomes:

$$E_{TP}(\omega_j) = \frac{\sum_k AC_{j,j-k}^{meas} \cdot E_{RP}(\omega_j - \Omega_k)}{\sum_k |E_{RP}(\omega_j - \Omega_k)|^2}$$

$$E_{RP}^*(\omega_j) = \frac{\sum_k AC_{j,j+k}^{meas} \cdot E_{TP}^*(\omega_j + \Omega_k)}{\sum_k |E_{TP}(\omega_j + \Omega_k)|^2} \quad (7.1)$$

The algorithm is performed with 50 iterations and the phases of the two fields are so retrieved. The spectra are directly acquired with the spectrometer by sending one waveform at a time. The result of the TP phase of the test pulse and the reference are shown in Fig. 7.7 b) and c) and show good agreement with the phase retrieved by means of a conventional SPIDER in Fig. 7.7 a). Although the behaviour

of the phase is similar in the three reconstructions, the differences are more evident in Fig 7.7 (b) than in Fig 7.7 (c), that is, the test pulse rather than the reference reconstruction. The reason might be that as stated before, even in this case the copies are not effectively sheared, and, since the phase, even if nearly flat, still presents features, which are not handled well when dealing with a varying pulse.

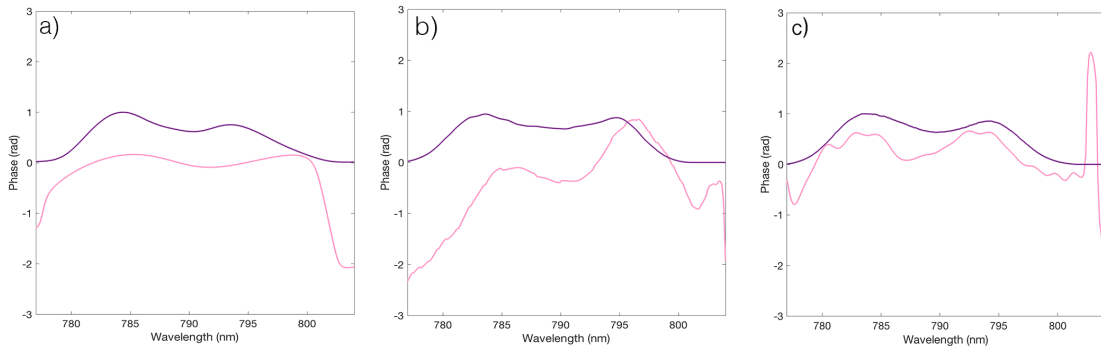


Fig. 7.7 Results: a) Normalized spectrum (purple) and spectral phase (pink) measured with a commercial SPIDER. b) Normalized spectrum (purple) and spectral phase (pink) of the test pulse obtained with a 2-field MICE retrieval c) Normalized spectrum (purple) and spectral phase (pink) of the reference pulse obtained with a 2-field MICE retrieval.

These results are the preliminary step to expand MICE to the XUV regime. The next step will involve using a similar setup, but with an amplification system after the DAZZLER so to add the capability of obtaining HHG, in order to characterize single harmonics with MICE. A further step would be to use the same device with a suitably large pulse as a reference so to characterize the full HHG beam by bridging the spectral-gaps for an XUV-SPICE implementation.

CONCLUSIONS

Finishing a hat, starting on a hat

Finishing a hat, look I made a hat

Where there never was a hat.

Sunday in the park with George, S. Sondheim

The concepts developed in this thesis provide an important metrology tool for the development of new light sources requiring higher pulses complexity and as such give good grounds for many applications. The most straightforward application for SPICE is in fact as diagnostic for coherent synthesis sources (as the one developed by HWU and those discussed in Chapter 2). The MICE algorithm opens to the possibility of characterizing more than one pulse at the same time without ambiguities: this can be particularly useful even in "standard" pulses characterization, in instances where there are strict power limitations and a prior characterization of an external reference is not favorable. The applications of MICE are virtually endless, as it is a very flexible characterization technique where the only requirement is to adopt a multi-shear approach. Some of the possibilities have recently been exploited by

our group and include spatial characterization of HHG [111] and the application of MICE to digital holographic microscopy [112].

8.1. SUMMARY

As sources for able to synthesise arbitrary light pulses develop, introducing novel techniques for their characterisation is key to their real employ for spectroscopy. My thesis aims at being a contribution towards this goal. In detail, I have tackled the longstanding issue of retrieving the relative phase of two well-separated spectral components with a new interferometric technique. This manuscript discusses the different routes that we have explored towards accomplishing our set goals. Based on the specific demands of our collaborator Prof.Reid concerning their coherent light source, we have devised an XFROG setup sensitive to low intensities to gather information on the components of their source. This has provided insights on the phase of their source, allowing to compress individual components and control the relative delays. The issue of retrieving the relative phase remained however elusive.

This has brought us to conceiving a radically different approach, by developing SPICE. This new technique has proven to be successful in the retrieval of the relative phase. This has been demonstrated with simulations, which have shown a high robustness to noise, and I have also reported of an experimental implementation, at least in a test-bed configuration.

SPICE success has been enabled by the development of a new algorithm, MICE. By granting the possibility of *mutually* characterising multiple fields, i.e. without making use of a known reference pulse, MICE allowed the reconstruction of the relative phase avoiding the necessity of a previously-measured broad reference.

REFERENCES

- [1] E. Ippen, C. Shank, and A. Dienes, "Passive mode locking of the cw dye laser," *Applied Physics Letters*, vol. 21, pp. 348–350, oct 1972.
- [2] L. E. Hargrove, R. L. Fork, and M. A. Pollack, "LOCKING OF He–Ne LASER MODES INDUCED BY SYNCHRONOUS INTRACAVITY MODULATION," *Applied Physics Letters*, vol. 5, pp. 4–5, jul 1964.
- [3] P. W. Smith, "Mode-Locking of Lasers," in *Proceedings of the IEEE* vol. 58 No. 9, 1970.
- [4] D. Polli, P. Altoè, O. Weingart, K. M. Spillane, C. Manzoni, D. Brida, G. Tomasello, G. Orlandi, P. Kukura, R. a. Mathies, M. Garavelli, and G. Cerullo, "Conical intersection dynamics of the primary photoisomerization event in vision," *Nature*, vol. 467, no. 7314, pp. 440–443, 2010.
- [5] O. M. Sarkisov, A. N. Petrukhin, F. E. Gostev, and A. A. Titov, "Control of elementary chemical reactions by femtosecond light pulses," *Quantum Electronics*, vol. 31, pp. 483–488, jun 2001.
- [6] d. D. von Linde and H. Schuler, "Breakdown threshold and plasma formation in femtosecond laser-solid interaction," *J. Opt. Soc. Am. B*, vol. 13, no. 1, pp. 216–222, 1996.
- [7] R. Ell, U. Morgner, F. X. Kärtner, J. G. Fujimoto, E. P. Ippen, V. Scheuer, G. Angelow, T. Tschudi, M. J. Lederer, A. Boiko, and B. Luther-Davies, "Generation of 5-fs pulses and octave-spanning spectra directly from a Ti:sapphire laser," *Optics Letters*, vol. 26, no. 6, p. 373, 2001.
- [8] I. D. Jung, F. X. Kärtner, N. Matuschek, D. H. Sutter, F. Morier-Genoud, G. Zhang, U. Keller, V. Scheuer, M. Tilsch, and T. Tschudi, "Self-starting 6.5-fs pulses from a Ti:sapphire laser," *Optics Letters*, vol. 22, no. 13, pp. 1009–1011, 1997.
- [9] M. Nisoli, S. De Silvestri, and O. Svelto, "Generation of high energy 10 fs pulses by a new pulse compression technique," *Applied Physics Letters*, vol. 68, no. 20, pp. 2793–2795, 1996.
- [10] M. Nisoli, S. De Silvestri, O. Svelto, R. Szipöcs, K. Ferencz, C. Spielmann, S. Sartania, and F. Krausz, "Compression of high-energy laser pulses below 5 fs.," *Optics letters*, vol. 22, no. 8, pp. 522–524, 1997.

- [11] O. D. Mucke, S. Alisauskas, A. J. Verhoef, A. Pugzlys, A. Baltuska, V. Smilgevicius, J. Pocius, L. Giniunas, R. Danielius, and N. Forget, "Self-compression of millijoule 1.5 μm pulses," *Optics Letters*, vol. 34, no. 16, pp. 2498–2500, 2009.
- [12] D. Brida, C. Manzoni, G. Cirimi, M. Marangoni, S. Bonora, P. Villoresi, S. De Silvestri, and G. Cerullo, "Few-optical-cycle pulses tunable from the visible to the mid-infrared by optical parametric amplifiers," *Journal of Optics*, vol. 12, p. 013001, jan 2010.
- [13] I. A. Walmsley and C. Dorrer, "Characterization of ultrashort electromagnetic pulses," *Advances in Optics and Photonics*, vol. 1, no. 2, p. 308, 2009.
- [14] R. Trebino, "Frequency-Resolved Optical Gating: The Measurement of Ultrashort Laser Pulses," 2000.
- [15] A. Monmayrant, S. Weber, and B. Chatel, "A newcomer's guide to ultrashort pulse shaping and characterization," *Journal of Physics B: Atomic, Molecular and Optical Physics*, vol. 43, no. 10, p. 103001, 2010.
- [16] S. Linden, H. Giessen, and J. Kuhl, "XFROG—a new method for amplitude and phase characterization of weak ultrashort pulses," *Physica Status Solidi (B)*, vol. 206, no. 1, pp. 119–124, 1998.
- [17] D. N. Fittinghoff, J. L. Bowie, J. N. Sweetser, R. T. Jennings, M. a. Krumbügel, K. W. Delong, R. Trebino, and I. a. Walmsley, "Measurement of the intensity and phase of ultraweak, ultrashort laser pulses," *Optics letters*, vol. 21, no. 12, pp. 884–886, 1996.
- [18] E. M. Kosik, A. S. Radunsky, I. A. Walmsley, and C. Dorrer, "Interferometric technique for measuring broadband ultrashort pulses at the sampling limit," *Optics letters*, vol. 30, no. 3, pp. 326–328, 2005.
- [19] P. Baum, S. Lochbrunner, and E. Riedle, "Zero-additional-phase SPIDER: full characterization of visible and sub-20-fs ultraviolet pulses," *Optics letters*, vol. 29, no. 2, pp. 210–212, 2004.
- [20] C. Dorrer, P. Londero, and I. a. Walmsley, "Homodyne detection in spectral phase interferometry for direct electric-field reconstruction," *Optics letters*, vol. 26, no. 19, pp. 1510–1512, 2001.
- [21] T. Witting, D. R. Austin, and I. A. Walmsley, "Ultrashort pulse characterization by spectral shearing interferometry with spatially chirped ancillae," *Optics express*, vol. 17, no. 21, pp. 18983–18994, 2009.
- [22] D. R. Austin, T. Witting, and I. A. Walmsley, "High precision self-referenced phase retrieval of complex pulses with multiple-shearing spectral interferometry," *Journal of the Optical Society of America B*, vol. 26, no. 9, p. 1818, 2009.
- [23] D. R. Austin, T. Witting, and I. A. Walmsley, "Resolution of the relative phase ambiguity in spectral shearing interferometry of ultrashort pulses," *Optics letters*, vol. 35, no. 12, pp. 1971–1973, 2010.

- [24] P. B. Corkum, "Plasma perspective on strong field multiphoton ionization," *Physical Review Letters*, vol. 71, no. 13, pp. 1994–1997, 1993.
- [25] F. Krausz and M. Ivanov, "Attosecond physics," *Reviews of Modern Physics*, vol. 81, no. 1, pp. 163–234, 2009.
- [26] P. M. Paul, E. S. Toma, P. Breger, G. Mullot, F. Auge, P. Balcou, H. G. Muller, and P. Agostini, "Observation of a Train of Attosecond Pulses from High Harmonic Generation," *Science*, vol. 292, no. 5522, pp. 1689–1692, 2001.
- [27] A. Wirth, M. T. Hassan, I. Grguras, J. Gagnon, A. Moulet, T. T. Luu, S. Pabst, R. Santra, Z. a. Alahmed, a. M. Azzeer, V. S. Yakovlev, V. Pervak, F. Krausz, and E. Goulielmakis, "Synthesized Light Transients," *Science*, vol. 334, no. 6053, pp. 195–200, 2011.
- [28] G. Sansone, E. Benedetti, F. Calegari, C. Vozzi, L. Avaldi, R. Flammini, L. Poletto, P. Villoresi, C. Altucci, R. Velotta, S. Stagira, S. De Silvestri, and M. Nisoli, "Isolated Single-Cycle Attosecond Pulses," *Science*, vol. 314, no. 5798, pp. 443–446, 2006.
- [29] E. J. Takahashi, P. Lan, O. D. Mücke, Y. Nabekawa, and K. Midorikawa, "Infrared two-color multicycle laser field synthesis for generating an intense attosecond pulse," *Physical Review Letters*, vol. 104, no. 23, 2010.
- [30] E. J. Takahashi, P. Lan, O. D. Mücke, Y. Nabekawa, and K. Midorikawa, "Attosecond nonlinear optics using gigawatt-scale isolated attosecond pulses," *Nature communications*, vol. 4, p. 2691, 2013.
- [31] T. Brabec and F. Krausz, "Intense few-cycle laser fields: Frontiers of nonlinear optics," *Reviews of Modern Physics*, vol. 72, no. 2, pp. 545–591, 2000.
- [32] S. Aktürk, X. Gu, P. Gabolde, and R. Trebino, "First-order spatiotemporal distortions of gaussian pulses and beams," *Springer Series in Optical Sciences*, vol. 132, no. 21, pp. 233–239, 2007.
- [33] Z. Bor, "Femtosecond pulse front tilt caused by angular dispersion," *Optical Engineering*, vol. 32, no. 10, p. 2501, 1993.
- [34] J. Hebling, "Derivation of the pulse front tilt caused by angular dispersion," *Optical and Quantum Electronics*, vol. 28, no. 12, pp. 1759–1763, 1996.
- [35] S. Akturk, X. Gu, E. Zeek, and R. Trebino, "Pulse-front tilt caused by spatial and temporal chirp," *Optics Express*, vol. 12, no. 19, pp. 4399–4410, 2004.
- [36] E. Hecht, "Optics," in *Optics*, 2002.
- [37] B. Tatian, "Fitting refractive-index data with the Sellmeier dispersion formula," *Applied optics*, vol. 23, no. 24, pp. 4477–4485, 1984.
- [38] I. A. Walmsley, L. Waxer, and C. Dorrer, "The role of dispersion in ultrafast optics," 2001.

- [39] R. L. Fork, O. E. Martinez, and J. P. Gordon, "Negative dispersion using pairs of prisms," *Opt. Lett.*, vol. 9, no. 5, pp. 150–152, 1984.
- [40] C. Manzoni, O. D. Mucke, G. Cirimi, S. Fang, J. Moses, S. W. Huang, K. H. Hong, G. Cerullo, and F. X. Kartner, "Coherent pulse synthesis: Towards sub-cycle optical waveforms," *Laser and Photonics Reviews*, vol. 9, no. 2, pp. 129–171, 2015.
- [41] E. B. Treacy, "Optical Pulse Compression With Diffraction Gratings," *IEEE Journal of Quantum Electronics*, vol. 5, no. 9, pp. 454–458, 1969.
- [42] R. Szipöcs, C. Spielmann, F. Krausz, and K. Ferencz, "Chirped multilayer coatings for broadband dispersion control in femtosecond lasers," *Optics Letters*, vol. 19, no. 3, p. 201, 1994.
- [43] F. X. Kärtner, N. Matuschek, T. Schibli, U. Keller, H. A. Haus, C. Heine, R. Morf, V. Scheuer, M. Tilsch, and T. Tschudi, "Design and fabrication of double-chirped mirrors," *Optics Letters*, vol. 22, no. 11, pp. 831–833, 1997.
- [44] E. A. Gibson, D. M. Gaudiosi, H. C. Kapteyn, R. Jimenez, R. Huff, C. Durfee, and J. Squier, "Efficient reflection grisms for pulse compression and dispersion compensation of femtosecond pulses," vol. 31, no. 22, pp. 3363–3365, 2006.
- [45] A. M. Weiner, "Ultrafast optical pulse shaping: A tutorial review," 2011.
- [46] A. M. Weiner, "Femtosecond pulse shaping using spatial light modulators," *Review of Scientific Instruments*, vol. 71, no. 5, p. 1929, 2000.
- [47] T. Binhammer, E. Rittweger, R. Ell, F. X. Kärtner, and U. Morgner, "Prism-based pulse shaper for octave spanning spectra," *IEEE Journal of Quantum Electronics*, vol. 41, no. 12, pp. 1552–1557, 2005.
- [48] Robert W. Boyd, "Nonlinear Optics," 2013.
- [49] F. Parmigiani, C. Finot, K. Mukasa, M. Ibsen, M. a. Roelens, P. Petropoulos, and D. J. Richardson, "Ultra-flat SPM-broadened spectra in a highly nonlinear fiber using parabolic pulses formed in a fiber Bragg grating," *Optics Express*, vol. 14, no. 17, p. 7617, 2006.
- [50] D. J. e. a. Jones, "Carrier-envelope phase control of femtosecond mode-locked lasers and direct optical frequency synthesis," *Science (New York, N.Y.)*, vol. 288, no. 5466, pp. 635–40, 2000.
- [51] U. Morgner, R. Ell, G. Metzler, T. R. Schibli, F. X. Kärtner, J. G. Fujimoto, H. A. Haus, and E. P. Ippen, "Nonlinear optics with phase-controlled pulses in the sub-two- cycle regime," *Physical Review Letters*, vol. 86, no. 24, pp. 5462–5465, 2001.
- [52] G. Krauss, S. Lohss, T. Hanke, A. Sell, S. Eggert, R. Huber, and A. Leitenstorfer, "Synthesis of a single cycle of light with compact erbium-doped fibre technology," 2010.

- [53] S.-W. Huang, G. Cirimi, J. Moses, K.-H. Hong, S. Bhardwaj, J. R. Birge, L.-J. Chen, E. Li, B. J. Eggleton, G. Cerullo, and F. X. Kärtner, "High-energy pulse synthesis with sub-cycle waveform control for strong-field physics," *Nature Photonics*, vol. 5, pp. 475–479, jul 2011.
- [54] R. A. McCracken, J. Sun, C. G. Leburn, and D. T. Reid, "Broadband phase coherence between an ultrafast laser and an OPO using lock-to-zero CEO stabilization," 2012.
- [55] H. P. Weber, "Method for pulsewidth measurement of ultrashort light pulses generated by phase-locked lasers using nonlinear optics," *Journal of Applied Physics*, vol. 38, no. 5, pp. 2231–2234, 1967.
- [56] J. A. Armstrong, "Measurement of picosecond laser pulse widths," *Applied Physics Letters*, vol. 10, no. 1, pp. 16–18, 1967.
- [57] K. Naganuma, K. Mogi, and H. Yamada, "General Method for Ultrashort Light Pulse Chirp Measurement," *IEEE Journal of Quantum Electronics*, vol. 25, no. 6, pp. 1225–1233, 1989.
- [58] R. Trebino and D. J. Kane, "Using phase retrieval to measure the intensity and phase of ultrashort pulses: frequency-resolved optical gating," *Journal of the Optical Society of America A*, vol. 10, no. 5, p. 1101, 1993.
- [59] D. J. Kane and R. Trebino, "Characterization of arbitrary femtosecond pulses using frequency-resolved optical gating," 1993.
- [60] D. J. Kane and R. Trebino, "Single-shot measurement of the intensity and phase of an arbitrary ultrashort pulse by using frequency-resolved optical gating," *Optics letters*, vol. 18, no. 10, pp. 823–825, 1993.
- [61] R. E. Berg and D. G. Stork, *The physics of sound*. Prentice-Hall, 1982.
- [62] L. Cohen and Leon, *Time-frequency analysis*. Prentice Hall PTR, 1995.
- [63] R. Trebino, K. W. DeLong, D. N. Fittinghoff, J. N. Sweetser, M. A. Krumbugel, B. A. Richman, and D. J. Kane, "Measuring ultrashort laser pulses in the time-frequency domain using frequency-resolved optical gating," *Review of Scientific Instruments*, vol. 68, no. 9, p. 3277, 1997.
- [64] K. W. DeLong, R. Trebino, and W. E. White, "Simultaneous recovery of two ultrashort laser pulses from a single spectrogram," *Journal of the Optical Society of America B*, vol. 12, no. 12, p. 2463, 1995.
- [65] J. Paye, M. Ramaswamy, J. G. Fujimoto, and E. P. Ippen, "Measurement of the amplitude and phase of ultrashort light pulses from spectrally resolved autocorrelation," *Optics letters*, vol. 18, no. 22, pp. 1946–8, 1993.
- [66] T. Tsang, M. Krumbugel, K. W. DeLong, D. N. Fittinghoff, and R. Trebino, "Frequency-resolved optical-gating measurements of ultrashort pulses using surface third-harmonic generation," *Optics letters*, vol. 21, no. 17, pp. 1381–3, 1996.

- [67] K. W. DeLong, R. Trebino, J. Hunter, and W. E. White, "Frequency-resolved optical gating with the use of second-harmonic generation," *Journal of the Optical Society of America B*, vol. 11, no. 11, p. 2206, 1994.
- [68] J. Paye, "How to measure the amplitude and phase of an ultrashort light pulse with an autocorrelator and a spectrometer," *IEEE Journal of Quantum Electronics*, vol. 30, no. 11, pp. 2693–2697, 1994.
- [69] G. Taft, a. Rundquist, M. M. Murnane, H. C. Kapteyn, K. W. DeLong, R. Trebino, and I. P. Christov, "Ultrashort optical waveform measurements using frequency-resolved optical gating," *Optics letters*, vol. 20, no. 7, pp. 743–745, 1995.
- [70] A. Baltuska, M. Pshenichnikov, and D. Wiersma, "Second-harmonic generation frequency-resolved optical gating in the single-cycle regime," *IEEE Journal of Quantum Electronics*, vol. 35, pp. 459–478, apr 1999.
- [71] Z. Penman, T. Schittkowski, W. Sleat, D. Reid, and W. Sibbett, "Experimental comparison of conventional pulse characterisation techniques and second-harmonic-generation frequency-resolved optical gating," *Optics Communications*, vol. 155, no. 4, pp. 297–300, 1998.
- [72] G. Taft, A. Rundquist, M. Murnane, I. Christov, H. Kapteyn, K. DeLong, D. Fittinghoff, M. Krumbugel, J. Sweetser, and R. Trebino, "Measurement of 10-fs laser pulses," *IEEE Journal of Selected Topics in Quantum Electronics*, vol. 2, no. 3, pp. 575–585, 1996.
- [73] B. Richman, K. DeLong, and R. Trebino, "Temporal characterization of the Stanford mid-IR FEL micropulses by "FROG"," *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, vol. 358, no. 1, pp. 268–271, 1995.
- [74] B. Kohler, J. Squier, K. W. DeLong, R. Trebino, V. V. Yakovlev, and K. R. Wilson, "Phase and intensity characterization of femtosecond pulses from a chirped-pulse amplifier by frequency-resolved optical gating," *Optics Letters*, vol. 20, p. 483, mar 1995.
- [75] D. J. Kane, G. Rodriguez, a. J. Taylor, and T. S. Clement, "Simultaneous measurement of two ultrashort laser pulses from a single spectrogram in a single shot," *Journal of the Optical Society of America B*, vol. 14, no. 4, p. 935, 1997.
- [76] A. K. Jain, *Fundamentals of digital image processing*. Englewood Cliffs NJ: Prentice Hall, 1989.
- [77] K. W. DeLong, D. N. Fittinghoff, R. Trebino, B. Kohler, and K. Wilson, "Pulse retrieval in frequency-resolved optical gating based on the method of generalized projections," *Optics letters*, vol. 19, no. 24, pp. 2152–2154, 1994.
- [78] E. Yudilevich, A. Levi, G. J. Habetler, and H. Stark, "Restoration of signals from their signed Fourier-transform magnitude by the method of generalized projections," *Journal of the Optical Society of America A*, vol. 4, no. 1, p. 236, 1987.

- [79] V. Wong and I. a. Walmsley, "Analysis of ultrashort pulse-shape measurement using linear interferometers.," *Optics letters*, vol. 19, no. 4, p. 287, 1994.
- [80] C. Dorrer and I. Kang, "Highly sensitive direct characterization of femtosecond pulses by electro-optic spectral shearing interferometry.," *Optics letters*, vol. 28, no. 6, pp. 477–479, 2003.
- [81] I. Kang, C. Dorrer, and F. Quochi, "Implementation of electro-optic spectral shearing interferometry for ultrashort pulse characterization.," *Optics letters*, vol. 28, no. 22, pp. 2264–2266, 2003.
- [82] C. Iaconis and I. a. Walmsley, "Spectral phase interferometry for direct electric-field reconstruction of ultrashort optical pulses.," *Optics letters*, vol. 23, no. 10, pp. 792–794, 1998.
- [83] M. Takeda, H. Ina, and S. Kobayashi, "Fourier-transform method of fringe-pattern analysis for computer-based topography and interferometry," *J. Opt. Soc. Am.*, vol. 72, pp. 156–160, jan 1982.
- [84] C. Dorrer and I. A. Walmsley, "Accuracy criterion for ultrashort pulse characterization techniques: application to spectral phase interferometry for direct electric field reconstruction," *Journal of the Optical Society of America B*, vol. 19, no. 5, p. 1019, 2002.
- [85] M. E. Anderson, L. E. E. de Araujo, E. M. Kosik, and I. A. Walmsley, "The effects of noise on ultrashort-optical-pulse measurement using SPIDER," *Applied Physics B*, vol. 70, no. 1, pp. S85–S93, 2000.
- [86] P. Londero, Anderson, M. E., Radzewicz, Czeslaw, Iaconis, Chris, Walmsley, and I. A., "Measuring ultrafast pulses in the near-ultraviolet using spectral phase interferometry for direct electric field reconstruction," *J. Mod. Opt.*, vol. 50, no. 2, pp. 179–184, 2003.
- [87] W. Kornelis, J. Biegert, J. W. G. Tisch, M. Nisoli, G. Sansone, C. Vozzi, S. De Silvestri, and U. Keller, "Single-shot kilohertz characterization of ultrashort pulses by spectral phase interferometry for direct electric-field reconstruction.," *Optics letters*, vol. 28, no. 4, pp. 281–283, 2003.
- [88] C. Dorrer, P. Londero, and I. a. Walmsley, "Homodyne detection in spectral phase interferometry for direct electric-field reconstruction.," *Optics letters*, vol. 26, no. 19, pp. 1510–1512, 2001.
- [89] C. Dorrer, E. M. Kosik, and I. A. Walmsley, "Spatio-temporal characterization of the electric field of ultrashort optical pulses using two-dimensional shearing interferometry," *Applied Physics B: Lasers and Optics*, vol. 74, no. SUPPL., 2002.
- [90] A. S. Radunsky, E. M. K. Williams, I. A. Walmsley, P. Wasylczyk, W. Wasilewski, A. B. U'Ren, and M. E. Anderson, "Simplified spectral phase interferometry for direct electric-field reconstruction by using a thick nonlinear crystal.," *Optics letters*, vol. 31, no. 7, pp. 1008–10, 2006.

- [91] A. S. Radunsky, S. P. Gorza, I. A. Walmsley, and P. Wasylczyk, "A compact spectral shearing interferometer for ultrashort pulse characterization," *Conference on Lasers and Electro-Optics and 2006 Quantum Electronics and Laser Science Conference, CLEO/QELS 2006*, vol. 32, no. 2, pp. 2006–2008, 2006.
- [92] A. S. Radunsky, I. A. Walmsley, S.-P. Gorza, and P. Wasylczyk, "Compact spectral shearing interferometer for ultrashort pulse characterization," *Optics Letters*, vol. 32, p. 181, jan 2007.
- [93] A. S. Wyatt, I. A. Walmsley, G. Stibenz, and G. Steinmeyer, "Sub-10 fs pulse characterization using spatially encoded arrangement for spectral phase interferometry for direct electric field reconstruction," *Optics Letters*, vol. 31, no. 12, p. 1914, 2006.
- [94] R. Morita, M. Hirasawa, N. Karasawa, S. Kusaka, N. Nakagawa, K. Yamane, L. Li, A. Suguro, and M. Yamashita, "Sub-5 fs optical pulse characterization," *Measurement Science and Technology*, vol. 13, no. 11, pp. 1710–1720, 2002.
- [95] M. Hirasawa, N. Nakagawa, K. Yamamoto, R. Morita, H. Shigekawa, and M. Yamashita, "Sensitivity improvement of spectral phase interferometry for direct electric-field reconstruction for the characterization of low-intensity femtosecond pulses," *Applied Physics B: Lasers and Optics*, vol. 74, no. SUPPL., pp. 225–229, 2002.
- [96] C. Bourassin-Bouchet, M. M. Mang, I. Gianani, and I. a. Walmsley, "Mutual interferometric characterization of a pair of independent electric fields.," *Optics letters*, vol. 38, no. 24, pp. 5299–302, 2013.
- [97] D. Keusters, H.-S. Tan, P. O'Shea, E. Zeek, R. Trebino, and W. S. Warren, "Relative-phase ambiguities in measurements of ultrashort pulses with well-separated multiple frequency components," *Journal of the Optical Society of America B*, vol. 20, no. 10, p. 2226, 2003.
- [98] B. Seifert and H. Stolz, "A method for unique phase retrieval of ultrafast optical fields," *Measurement Science and Technology*, vol. 20, p. 015303, jan 2009.
- [99] W. P. de Boeij, M. S. Pshenichnikov, and D. a. Wiersma, "Heterodyne-detected stimulated photon echo: applications to optical dynamics in solution," *Chemical Physics*, vol. 233, no. 2-3, pp. 287–309, 1998.
- [100] K. Sundermann and R. de Vivie-Riedle, "Extensions to quantum optimal control algorithms and applications to special problems in state selective molecular dynamics," *J. Chem. Phys.*, vol. 110, no. 4, pp. 1896+, 1999.
- [101] B. Seifert, H. Stolz, and M. Tasche, "Nontrivial ambiguities for blind frequency-resolved optical gating and the problem of uniqueness," *Journal of the Optical Society of America B*, vol. 21, no. 5, p. 1089, 2004.
- [102] A. Hause, H. Hartwig, B. Seifert, H. Stolz, M. Bohm, and F. Mitschke, "Phase structure of soliton molecules," *Physical Review A*, vol. 75, no. 6, p. 063836, 2007.

- [103] R. A. McCracken, I. Gianani, A. S. Wyatt, and D. T. Reid, "Multi-color carrier-envelope-phase stabilization for high-repetition-rate multi-pulse coherent synthesis.," *Optics letters*, vol. 40, pp. 1208–11, apr 2015.
- [104] T. I. Ferreiro, J. Sun, and D. T. Reid, "Frequency stability of a femtosecond optical parametric oscillator frequency comb," *Optics Express*, vol. 19, no. 24, p. 24159, 2011.
- [105] D. C. Edelstein, E. S. Wachman, and C. L. Tang, "Broadly tunable high repetition rate femtosecond optical parametric oscillator," *Applied Physics Letters*, vol. 54, no. 18, pp. 1728–1730, 1989.
- [106] Y. Kobayashi, K. Torizuka, A. Marandi, R. L. Byer, R. A. McCracken, Z. Zhang, and D. T. Reid, "Femtosecond optical parametric oscillator frequency combs," *Journal of Optics*, vol. 17, no. 9, p. 094010, 2015.
- [107] M. M. Mang, C. Bourassin-Bouchet, and I. A. Walmsley, "Simultaneous spatial characterization of two independent sources of high harmonic radiation," *Optics Letters*, vol. 39, p. 6142, nov 2014.
- [108] P. B. Corkum and F. Krausz, "Attosecond science," *Nature Physics*, vol. 3, pp. 381–387, jun 2007.
- [109] A. S. Wyatt, T. Witting, A. Schiavi, D. Fabris, J. P. Marangos, J. W. G. Tisch, and I. A. Walmsley, "Attosecond sampling of arbitrary optical waveforms," *2013 Conference on Lasers and Electro-Optics Europe and International Quantum Electronics Conference, CLEO/Europe-IQEC 2013*, vol. 3, no. 3, 2013.
- [110] Y. Mairesse, O. Gobert, P. Breger, H. Merdji, P. Meynadier, P. Monchicourt, M. Perdrix, P. Salières, and B. Carré, "High harmonic XUV spectral phase interferometry for direct electric-field reconstruction," *Physical Review Letters*, vol. 94, no. 17, pp. 1–4, 2005.
- [111] M. M. Mang, C. Bourassin-Bouchet, and I. A. Walmsley, "Simultaneous spatial characterization of two independent sources of high harmonic radiation," *Optics Letters*, vol. 39, no. 21, p. 6142, 2014.
- [112] P. N. Anderson, I. Gianani, F. Wiegandt, K. Lloyd, M. M. Mng, A. Schiavi, K. O'Keffe, S. M. Hooker, and I. A. Walmsley, "Reference retrieval in digital holographic microscopy," *submitted to Optics Letters*, 2017.

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I've heard it say, that people come into our life
For a reason, bringing something we must learn
And we are lead to those who help us most to grow
If we let them, and we help them in return

Wicked, Stephen Schwartz

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