

ALBERT-LUDWIGS-UNIVERSITÄT FREIBURG

Attosekunden- und Starkfeldphysik am Physikalischen Institut

Bachelor Thesis

**Construction of an Attosecond Interferometer for
Two-Color Photoionization Experiments**



*A thesis submitted in fulfillment of the requirements
for the degree of Bachelor of Science in Physics.*

Author:

Víctor Delgado Pérez

Supervisor:

Prof. Dr. Giuseppe Sansone

March 4th - June 4th, 2024. Freiburg im Breisgau.

Kurzzusammenfassung

Bei Photoionisationsexperimenten werden Elektronen durch die Absorption hochenergetischer Photonen aus ihren Atomen herausgelöst. Sie sind ein ideales Instrument zur Untersuchung von Licht-Materie-Wechselwirkungen. Diese Arbeit widmet sich der Optimierung eines Attosekunden-Interferometers, das Zwei-Farben Photoionisationsexperimente in einer Anregungs-Abfrage-Konfiguration (pump-probe) ermöglicht. Ein hochenergetischer Laserpuls (die Anregung) wird verwendet, um ein bestimmtes Gas anzuregen, während ein anderer Laserpuls (die Abfrage) die Entwicklung dieses Prozesses in Abhängigkeit der Zeit überwacht. Zunächst wurde ein Ein-Farbfeld-Photoionisationsexperiment durchgeführt, bei dem ein Attosekunden-Pulszug im extrem-ultravioletten Bereich (XUV) zunächst mittels High Harmonic Generation (HHG) erzeugt wurde. Nach der Beschreibung seines Spektrums wurde er zur Ionisierung eines Argon-Atomstrahls eingesetzt. Die Photoionisation wurde mit einem Velocity-Map-Imaging-Spektrometer (VMI) untersucht, und die Eigenschaften des entsprechenden Photoelektronenspektrums wurden aufgezeichnet. Der zweiten Schritt war ein Zwei-Farben-Experiment, bei dem der zur Ionisierung verwendete Pumpstrahl mit dem eines kommerziell hergestellten titandotierten Saphir-Lasersystems im Nahinfrarot Bereich (near-infrared, NIR) und Femtosekunden-Ausgang überlagert wurde. Nach dem Bau eines Hilfsteleskops und der Ergänzung des ursprünglichen Aufbaus konnten schließlich Seitenbänder im Photoelektronenspektrum beobachtet werden, die auf die Wechselwirkung der Photoelektronen mit dem NIR-Feld zurückzuführen sind. Daneben wurde auch der Ausgang eines optischen parametrischen Verstärkers (optical parametric amplifier, OPA) charakterisiert, um ihn in Zukunft als Abfrage zu verwenden. Dabei wurden Leistung, Spektrum und Pulsdauer in beiden seiner Stufen analysiert, ebenso wie die Intensitätsstabilität des verstärkten Ausgangs. Die Eigenschaften des Spektrums nach der Verstärkung durch zwei verschiedene nichtlineare Kristalle mit unterschiedlichen Dicken wurden ebenfalls analysiert. Zur Messung der CEP-Fluktuationen wurde ein f-zu-2f-Interferometer gebaut und ein Fourier-basierter Algorithmus implementiert.

Abstract

Photoionization experiments study the mechanisms by which electrons are pulled apart from their atoms after absorbing high-energy photons, and they represent ideal tools to investigate light-matter interactions. This thesis has been dedicated to the optimization of an attosecond interferometer which allows for two-color photoionization experiments under a pump-probe configuration. A high-energy laser pulse (the pump) is used to excite a certain gas, while another laser pulse (the probe) monitors the evolution of this process as a function of time. To start with, a one color-field photoionization experiment was performed, where an attosecond pulse train in the extreme-ultraviolet range (XUV) was first created via high harmonic generation (HHG). After describing its spectrum, it was sent to ionize an atomic beam of argon. Photoionization was investigated using a velocity-map imaging (VMI) spectrometer, and the properties of the corresponding photoelectron spectrum were noted. A two color-field experiment followed, where the XUV radiation was recombined with the near-infrared (NIR) femtosecond output of a commercially produced Titanium-doped Sapphire laser system. After the construction and addition of an auxiliary telescope to the original setup, sidebands in the photoelectron spectrum were finally observed as a result of the interaction of photoelectrons with the NIR field. To conclude, the output of an optical parametric amplifier (OPA) was characterized with the purpose of using it as the probe in future two color-field experiments: power, spectrum and pulse duration were analysed at both of its stages, as well as the intensity stability of the amplified output. The properties of the spectrum after amplification by two distinct nonlinear crystals with different thicknesses were also analysed. To measure the CEP fluctuations, an f-to-2f interferometer was built and a Fourier-based algorithm was implemented.

Contents

Kurzzusammenfassung	i
Abstract	iii
List of figures	viii
List of tables	ix
Introduction	1
1 Theoretical Background	3
1.1 Optical Parametric Amplification	3
1.1.1 Optical Parametric Amplification for Monochromatic Waves . .	4
1.1.2 Phase-matching	8
1.1.3 Design criteria of ultrafast OPAs	10
1.2 White Light Generation	13
1.2.1 Optical Kerr Effect	13
1.3 Velocity Map Imaging	18
1.3.1 Traditional setup and working principle	19
1.3.2 Image Reconstruction	21
1.4 Reconstruction of Attosecond Beating By Interference of Two-photon Transitions	23
1.4.1 Motivation	23
1.4.2 Description	24
2 Beamline	29
2.1 The Ti:Sa laser system	30
2.2 The NOPA	31
2.3 The Attosecond Interferometer	33
2.4 The VMI spectrometer	36
2.5 The XUV spectrometer	37
3 Measurements, Analysis & Results	39
3.1 One color-field experiment: XUV only	40
3.1.1 Optimization and characterization of the harmonics	40
3.1.2 VMI measurements	42
3.2 Two color-field experiment: XUV + NIR	46
3.2.1 Construction of an auxiliar telescope	46
3.2.2 VMI measurements	48
3.3 Optimization of the NOPA	51
3.3.1 Power, spectrum & Fourier limit	51
3.3.2 Intensity stability	57
3.3.3 CEP stability	59

4 Summary and Outlook

67

5 Appendix

- 5.1 Nonlinear optics
- 5.2 Anisotropic media

Bibliography

Declaration

Acknowledgements

List of Figures

1.1	Sketch of the working principle of an optical parametric amplifier (OPA): undulatory point of view	3
1.2	Sketch of the working principle of an OPA: corpuscular point of view	4
1.3	Positive feedback of the idler to the amplification process	7
1.4	Illustration of the temporal walk-off of two light pulses	11
1.5	Definition of carrier-envelope phase (CEP)	12
1.6	Illustration of the self-focusing effect	15
1.7	Illustration of self-phase modulation (SPM)	16
1.8	Illustration of the self-steepening effect	16
1.9	Example of supercontinuum	17
1.10	Eppink-Parker-Desing of the velocity map imaging (VMI) spectrometer	19
1.11	Front and side views of a pair of microchannel plates (MCPs)	20
1.12	Comparison of the resolution of the photo-fragment imaging (PFI) and VMI techniques	21
1.13	Schematic of a cylindrically symmetric 3D distribution and its 2D pro- jection	22
1.14	Example of PES and corresponding energy level diagram in a one color- field experiment	25
1.15	Example of PES and corresponding energy level diagram in a two color- field experiment	26
2.1	Schematic overview of the beamline	29
2.2	Diagram of the different stages of the Titanium-doped Sapphire laser system	30
2.3	Illustration of the chirped pulse amplification (CPA) technique	31
2.4	Sketch of the home-built non-collinear OPA (NOPA)	32
2.5	Sketch of the attosecond interferometer	34
2.6	Sketch of the VMI spectrometer	36
3.1	Transmission curve of the 99 nm—thick aluminum filter used to block the near-infrared (NIR) field	40
3.2	Reflection efficiency of the grating in the XUV spectrometer and har- monic spectrum	41
3.3	Sketch of the typical spectrum resulting from the HHG process	42
3.4	Raw VMI measurement in the one color-field experiment	43
3.5	Abel-inverted image in the one color-field experiment	44
3.6	Normalized PES of the one color-field experiment	45
3.7	Sketch of the auxiliar telescope used to reduce the size of the beam in the two color-field experiment	47
3.8	Raw VMI measurement in the two color-field experiment	48

3.9	Abel-inverted image in the two-color field experiment	49
3.10	Normalized PES of the two color-field experiment	49
3.11	Spectrum and Fourier limit of the signal after the first NOPA amplification stage	52
3.12	Spectrum and Fourier limit of the signal after the second NOPA amplification stage using a 1 mm-thick BBO nonlinear crystal	53
3.13	Spectrum and Fourier limit of the signal after the second NOPA amplification stage using a 1.4 mm-thick BBO nonlinear crystal	53
3.14	Sketch of the FROG setup	55
3.15	Qualitative illustration of the working principle of FROG and measurement of the pulse duration of the NOPA signal	56
3.16	Comparison between the intensity stability of two consecutive NOPA shots	57
3.17	Peak intensity stability of the signal over 4 minutes	58
3.18	Overall intensity stability of the signal over 4 minutes	59
3.19	Schematic of the working principle of an f-to-2f interferometer: frequency combs	61
3.20	Sketch of the home-built f-to-2f interferometer used to measure the CEP fluctuations	61
3.21	CEP stability: interference fringes over 83 minutes	63
3.22	Intensity heatmaps of the interference fringes	63
3.23	Fourier Transform Spectral Interferometry (FTSI): summary of procedure	65
3.24	CEP drift measurement: phase as a function of acquisition time	65
4.1	Energy level diagram of the two color-field experiment when the NOPA output is used as the probe	69
5.1	Breakdown of the harmonic approximation under strong fields	
5.2	Illustration of the phase-matching condition	
5.3	Schematic of the propagation of an electromagnetic wave inside a uniaxial medium	

List of Tables

1.1	Types of uniaxial crystals and possible polarization configurations for each.	9
3.1	Energies of detected harmonics in the VMI spectrometer and kinetic energies of the corresponding photoelectrons	43
3.2	Total absorption cross section of argon atoms as a function of energy .	44
3.3	Estimated absorption cross sections for harmonics 11 th and 13 th , calculated using a linear interpolation.	45
3.4	Initial measurements of the probe beam power at different stages of the attosecond interferometer	46
3.5	Measurements of the probe beam power at different stages of the attosecond interferometer after the installation of the telescope	48
5.1	First-, second- and third-order nonlinear effects.	

Introduction

A full and complete understanding of the Universe we live in involves not only the description of the huge galaxies that surround us, but also the characterization of the tiny atoms they (and we) are made of. In order to investigate how they are formed and how they interact with other atoms and molecules, it is necessary to analyse the incredibly fast electron dynamics that take place inside them. Progress in ultrafast optics during the last twenty years has opened the possibility of inspecting these phenomena with unprecedented time resolution, in the order of attoseconds ($1 \text{ as} = 10^{-18} \text{ s}$) [1]. A whole new field of research with multiple applications in Chemistry, Solid State Physics, Computational Science and Astrophysics, to name a few, has emerged.

A very effective method to study electron dynamics is through photon-matter interactions, where an ideally unperturbed atom is excited with light of well-known properties. If the incoming photons have enough energy, electrons are pulled apart from their atoms according to the so-called *photoelectric effect* described by Einstein in 1905 [2]. These *photoelectrons* are said to be *photoionized*, and can be detected and studied with spectrometers. Combining photons of different energies (“colors”) in the ionization process opens the possibility of studying more complex interactions.

This thesis has been focused on the optimization of an attosecond interferometer which enables the realization of two-color photoionization experiments. In general terms, a high-energy laser pulse (the pump) is used to excite a certain gas target, while a weaker one (the probe) is then recombined and used to monitor the temporal evolution of the interaction. More specifically, an attosecond pulse train created via high harmonic generation in the extreme ultraviolet range is employed to ionize an atomic beam of argon. After recombination with the near-infrared electric field of a commercially produced Titanium-doped Sapphire (Ti:Sa) laser system, photoelectrons’ kinetic energy is measured with a velocity-map imaging (VMI) spectrometer. Additionally, the output of an optical parametric amplifier (OPA) has been characterized with the purpose of using it in further two color-field photoionization experiments. This device transfers part of the energy of the Ti:Sa output, centered at wavelength $\lambda \approx 780 \text{ nm}$, to a weaker beam centered at $\lambda \approx 1200 \text{ nm}$.

Chapter 1 is dedicated to the discussion of important theoretical concepts involving the experimental work done in the thesis. Particularly, optical parametric amplification as a second-order nonlinear effect, white light generation as a third-order nonlinear effect, the Physics lying behind the VMI spectrometer used to detect the photoelectrons in the photoionization experiments, and the “Reconstruction of Attosecond Beating By Interference of Two-photon Transitions” (RABBITT) technique. It was originally conceived for the reconstruction of attosecond pulse trains [1], and it is nowadays a powerful tool for studying photoionization processes.

Chapter 2 continues with the description of the experimental setup used in the project: the beamline. This includes the commercially produced Ti:Sa laser system, the home-built non-collinear optical parametric amplifier, the attosecond interferometer, the VMI spectrometer and the XUV spectrometer.

The experimental procedure, measurements and analysis of the acquired data make up Chapter 3. First, a one color-field photoionization experiment was performed. The harmonic spectrum was measured with an XUV spectrometer and the photoelectron spectrum after ionization of an atomic beam of argon was extracted from the VMI measurements. Then, a two color-field experiment was conducted, where the harmonics were recombined with the output of the Ti:Sa laser system. For this, an auxiliary telescope was built and installed in the setup. Later, power, spectrum, pulse duration and intensity stability of the output of the OPA were characterized. The properties of the spectrum after amplification by two distinct BaB_2O_4 (“beta-barium-borate”) nonlinear crystals with different thicknesses was also compared. To measure the CEP fluctuations, an f-to-2f interferometer was built, and the Fourier Transform Spectral Interferometry (FTSI) algorithm was implemented.

To conclude with the main body of the thesis, a brief summary of the obtained results and an outlook of possible improvements and upcoming experiments are given in Chapter 4.

An Appendix with a more detailed description of nonlinear optics can be found after Chapter 4. It could be useful for the reader in order to understand how nonlinear crystals are used in OPAs to maximize the efficiency of the process via phase-matching.

The declaration of authorship and originality of the thesis, as well as the acknowledgements to all who contributed to this work in a direct or in an indirect way, closes this document.

Chapter 1

Theoretical Background

1.1 Optical Parametric Amplification

Optical Parametric Amplifiers (OPAs) are laser systems which make use of second-order nonlinear effects with the purpose of transferring energy from a very intense *pump* pulse with a certain (angular) frequency ω_3 to a much weaker *signal* pulse with a variable frequency $\omega_1 < \omega_3$ ¹, giving birth in this process to an *idler* with frequency $\omega_2 = \omega_3 - \omega_1$, which arises from energy conservation.

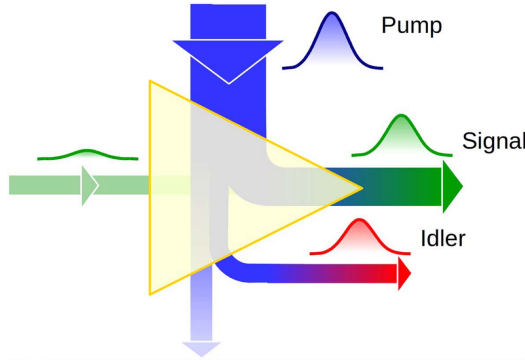


Figure 1.1: Basic sketch of an OPA. Taken and adapted from [3].

They constitute an essential tool in ultrafast optics with plenty of applications in multiple other fields as well. Their importance relies on the fact that not only they can be tuned to amplify a wide range of possible frequencies, but that they can be designed to amplify all these possible frequencies *at once*, thus acting as broadband amplifiers and accomplishing very short light pulses. Unlike classical optical amplifiers, their gain bandwidth is not limited by the linewidth of an electronic transition, but rather by the possibility of satisfying the phase-matching condition over a wide range of frequencies.

A full description of their working principle follows in the next sections. The reader will find an overview of nonlinear optics and birefringent materials (of utmost importance in this context) in the **Appendix**. Here, optical parametric amplification will be discussed for monochromatic waves in the first place. Subsequently, conditions for

¹The lower limit is technically determined by the parasitic absorption of the nonlinear crystal. Further details in the next pages.

phase-matching will be specified and the importance of the idler, highlighted. Finally, some technical criteria which should be taken into account in order to build ultrafast OPAs will be explained.

1.1.1 Optical Parametric Amplification for Monochromatic Waves

Since there is not a classical laser active medium able to provide acceptable gains over a broad frequency range from the mid-infrared to the visible, frequency tunability must be achieved by second-order nonlinear effects [3]. Optical parametric amplification can be understood from a corpuscular point of view, as illustrated in **Figure 1.2**: a *pump photon* of frequency ω_3 is first absorbed by a virtual level of the material². Then, a *signal photon* of frequency ω_1 stimulates the decay to an intermediate energy state and gets amplified; and finally, the ground state is reached by the release of a third *idler photon* of frequency ω_2 carrying the rest of the energy. Optical parametric amplification is therefore a *difference frequency generation* (DFG) process.

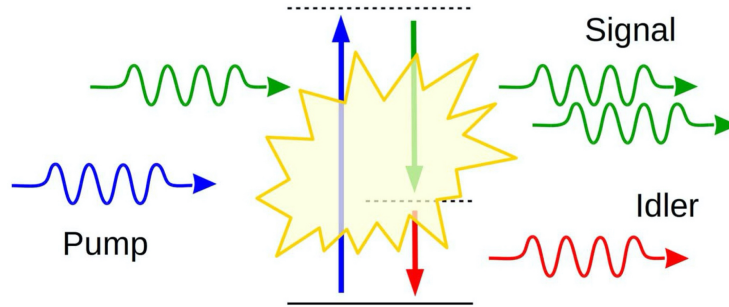


Figure 1.2: Corpuscular interpretation of the Optical Parametric Amplification process. Taken and adapted from [3].

It is important to note that since the response of the medium is instantaneous, the interaction will only occur if both the pump and signal photons cross the medium together [3]. Because of the second-order nonlinear nature of the process, this medium must be non-centrosymmetric, i.e. must have a non-zero second-order susceptibility $\chi^{(2)}$. Furthermore, since the collision probability increases with the number of photons, very intense beams are required.

According to the Fourier Theorem, a light pulse is a superposition of monochromatic waves. Assuming that all of the three interacting fields are plane waves propagating in the same z-direction, the resulting time-dependent electric field is:

²Virtual levels are not eigenstates of the atom; instead, they are regarded as eigenstates of the combined system “atom + photon”. Lifetimes associated with these levels are negligible compared to those of the eigenstates of the atom itself: according to the Uncertainty Principle, population can reside in a virtual level for a time interval of the order of $\hbar/\delta E$, where δE is the energy difference between the virtual level and the nearest “real” level. In other words, the closer the virtual state is to an actual eigenstate of the atom, the longer the lifetime of the virtual state. In fact, this is one of the main differences between *parametric* and *nonparametric* processes: while the former deal with virtual levels in which energy cannot be dissipated by radiative recombinations and therefore photon energy is totally conserved, the latter does involve the transfer of population from one real level to another, leading to heat dissipation and partial absorption of the photon energy [4,5].

$$E(z, t) = \frac{1}{2}[A_1(z)e^{i(\omega_1 t - k_1 z)} + A_2(z)e^{i(\omega_2 t - k_2 z)} + A_3(z)e^{i(\omega_3 t - k_3 z)}] + \text{c.c.}, \quad (1.1)$$

where the amplitudes A_i are complex and do not depend on time due to the monochromatic regime and $k_j = 2\pi/\lambda_j$ ($j = 1, 2, 3$) is the wavenumber. When an electromagnetic wave propagates through a medium, its associated electric field induces a force on the electrons, giving rise to a polarization P which can in turn affect the incident wave. This response is linear at low and moderate electric fields ($< 10^8$ V/m), and it is well described by the harmonic potential approximation of the Lorentz Model. However, under the influence of stronger electric fields, high-order terms appear in the Hook-type equations of motion. The anharmonicity of the potential becomes relevant and a perturbative treatment is necessary. Thereby, in the most general case, the polarization can be expressed as the sum of a linear contribution, $P_L(t)$, and a nonlinear one, $P_{NL}(t)$:

$$P(t) = P_L(t) + P_{NL}(t) = \underbrace{\varepsilon_0 \chi E(t)}_{P_L(t)} + \underbrace{\varepsilon_0 [\chi^{(2)} E^2(t) + \chi^{(3)} E^3(t) + \dots]}_{P_{NL}(t)}, \quad (1.2)$$

where ε_0 is the vacuum permittivity and $\chi^{(n)}$ ($n = 2, 3, \dots$) is the n th-order susceptibility tensor of rank $(n+1)$ of the dielectric medium. In order to account for the second-order nonlinear response of the medium, it is useful to define the so-called *effective nonlinear optical coefficient*: $d_{\text{eff}} = \chi^{(2)}/2$. Neglecting third and higher-order corrections, the nonlinear term can be written as [3]

$$P_{NL}(z, t) = 2\varepsilon_0 d_{\text{eff}} E^2(z, t) \quad (1.3)$$

In this equation, $E^2(z, t)$ will be given by the square of Eq.(1.1). Doing so, one can verify that the nonlinear second-order polarization contains components at various combinations of frequencies: second harmonics ($2\omega_1, 2\omega_2, 2\omega_3$), sums ($\omega_1 + \omega_2, \omega_1 + \omega_3, \omega_2 + \omega_3$) and differences ($\omega_3 - \omega_1, \omega_3 - \omega_2, \omega_2 - \omega_1$). Since OPA is a process which involves all ω_1 , ω_2 and ω_3 frequencies, it can be assumed that only those nonlinear interactions involving the three frequencies will efficiently take place:

$$\omega_1 + \omega_2 (= \omega_3) \qquad \omega_3 - \omega_1 (= \omega_2) \qquad \omega_3 - \omega_2 (= \omega_1), \quad (1.4)$$

With this, the nonlinear polarization term becomes

$$\begin{aligned} P_{NL}(z, t) = & \varepsilon_0 d_{\text{eff}} A_2^*(z) \cdot A_3(z) \cdot e^{i[\omega_1 t - (k_3 - k_2)z]} \\ & + \varepsilon_0 d_{\text{eff}} A_1^*(z) \cdot A_3(z) \cdot e^{i[\omega_2 t - (k_3 - k_1)z]} \\ & + \varepsilon_0 d_{\text{eff}} A_1^*(z) \cdot A_2(z) \cdot e^{i[\omega_3 t - (k_1 + k_2)z]} + \text{c.c.}, \end{aligned} \quad (1.5)$$

Deriving twice with respect to time:

$$\begin{aligned} \frac{\partial^2 P_{NL}(z, t)}{\partial t^2} = & -\varepsilon_0 d_{\text{eff}} \omega_1^2 A_2^*(z) \cdot A_3(z) \cdot e^{i[\omega_1 t - (k_3 - k_2)z]} \\ & - \varepsilon_0 d_{\text{eff}} \omega_2^2 A_1^*(z) \cdot A_3(z) \cdot e^{i[\omega_2 t - (k_3 - k_1)z]} \\ & - \varepsilon_0 d_{\text{eff}} \omega_3^2 A_1(z) \cdot A_2(z) \cdot e^{i[\omega_3 t - (k_1 + k_2)z]} + \text{c.c.}, \end{aligned} \quad (1.6)$$

and inserting this result into the the Maxwell's Equations written in the scalar approximation yields

$$\frac{\partial^2 E(z, t)}{\partial z^2} - \mu_0 \frac{\partial^2 D(z, t)}{\partial t^2} = \mu_0 \frac{\partial^2 P_{NL}(z, t)}{\partial t^2}, \quad (1.7)$$

where μ_0 is the magnetic permeability of vacuum and $D(z, t) = \varepsilon_0 E(z, t) + P_L(z, t)$ is the electric displacement field. Assuming local plane waves, it is possible now to apply the *slowly varying envelope approximation*:

$$\left| \frac{\partial^2 A}{\partial z^2} \right| \ll \left| \frac{\partial A}{\partial z} \right|, \quad (1.8)$$

and after splitting in components, the coupled wave equations are obtained:

$$\begin{cases} \frac{\partial A_1}{\partial z} = -i\sigma_1 A_2^* A_3 \cdot e^{-i\Delta k} \\ \frac{\partial A_2}{\partial z} = -i\sigma_2 A_1^* A_3 \cdot e^{-i\Delta k} \\ \frac{\partial A_3}{\partial z} = -i\sigma_3 A_1 A_2 \cdot e^{i\Delta k} \end{cases} \quad (1.9)$$

where $\sigma_j = d_{\text{eff}}\omega_j/cn_j$ (with c the speed of light in vacuum and $n_j = \sqrt{\varepsilon_{rj}} = \sqrt{1 + \chi(\omega_j)}$ the refractive index of the material) and $\Delta k = k_3 - k_2 - k_1$ is the so-called *phase mismatch*, representing the flow of energy between the three involved beams. The first two equations are formally identical, which suggests that the fields at ω_1 (signal) and ω_2 (idler) play the same role and can be interchanged. Therefore, the last expression of Eq.(1.4) is equivalent to the second one, which means that two processes can essentially take place:

1. *Sum frequency generation* (SFG), where the input fields are A_1 and A_2 at frequencies ω_1 and ω_2 , respectively, producing A_3 at $\omega_3 = \omega_1 + \omega_2$.
2. *Difference frequency generation*, where the input fields are A_3 and A_1 (or A_2) at frequencies ω_3 and ω_1 (or ω_2), respectively, producing A_2 (or A_1) at $\omega_2 = \omega_3 - \omega_1$ (or $\omega_1 = \omega_3 - \omega_2$). Under the assumption of an initially much stronger pump field, $A_3(z=0) \gg A_1(z=0)$, optical parametric amplification will occur³.

The detailed solution of the coupled equations (1.9) goes beyond the scope of this project; nevertheless it can be simplified under certain conditions:

Assuming first that the pump field stays approximately constant during the interaction ($A_3 \approx \text{const.}$) and that before the interaction there is no idler beam ($A_2(z=0) = 0$), the signal evolution along the nonlinear medium is described by [3]:

$$\frac{\partial^2 A_1}{\partial z^2} = -i\Delta k \frac{\partial A_1}{\partial z} + \Gamma^2 A_1, \quad \text{with} \quad \begin{cases} I_3 = n_3 \varepsilon_0 c \langle E(t)^2 \rangle = n_3 \varepsilon_0 c \cdot \frac{1}{2} |A_3|^2 \\ \Gamma^2 = \frac{2d_{\text{eff}}^2 \omega_1 \omega_2}{c^3 \varepsilon_0 n_1 n_2 n_3} I_3 \end{cases} \quad (1.10)$$

³Note however that the frequency relationship between the three fields is the same than in SFG, so both processes are essentially equivalent; the signature of each mechanism is simply the initial interacting fields.

where I_3 is the intensity of the pump, and n_1 , n_2 and n_3 are the refractive indexes for the signal, idler and pump (since they have different frequencies), respectively.

After a certain length L of interaction within the crystal, the signal and idler intensities are:

$$\begin{cases} I_1(L) = I_1(0) \left\{ 1 + \left[\frac{\Gamma}{g} \sinh(gL) \right]^2 \right\} \\ I_2(L) = I_1(0) \frac{\omega_2}{\omega_1} \left[\frac{\Gamma}{g} \sinh(gL) \right]^2 \end{cases}, \quad \text{with} \quad g = \sqrt{\Gamma^2 - \frac{\Delta k^2}{4}} \quad (1.11)$$

g is the so-called *small-signal gain*. The parametric gain $G(L)$ can be defined as

$$G(L) = \frac{I_1(L)}{I_1(0)} = \left\{ 1 + \left[\frac{\Gamma}{g} \sinh(gL) \right]^2 \right\} \quad (1.12)$$

As expected from the definition of g , it is maximized when $\Delta k = 0$, which is the so-called *phase-matching condition*. When $gL \gg 1$, that is, in the large-gain limit, Eqs.(1.11) and (1.12) can be approximated by

$$\begin{cases} I_1(L) \simeq I_1(0) \left(\frac{\Gamma}{g} \right)^2 \frac{e^{2gL}}{4} \\ I_2(L) \simeq I_1(0) \frac{\omega_2}{\omega_1} \left(\frac{\Gamma}{g} \right)^2 \frac{e^{2gL}}{4} \end{cases} \Rightarrow G(L) \simeq \left(\frac{\Gamma}{g} \right)^2 \frac{e^{2gL}}{4} \quad (1.13)$$

which shows how the gain grows exponentially with the interaction distance. A very important remark is that this is not only true for the signal, but also for the idler. Given that that one is mainly interested in amplifying the signal, the idler could initially be regarded as an unavoidable source of lost energy. However, it actually plays a vital role in the amplification process, acting as a positive feedback to the system as illustrated in **Figure 1.3**. Hence, if the idler is absorbed or depleted, the gain of the OPA gets drastically reduced.

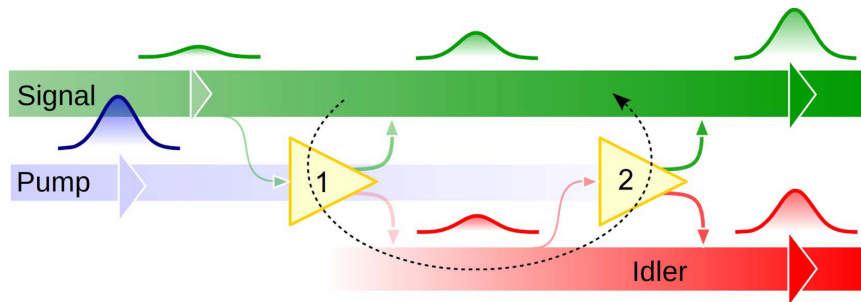


Figure 1.3: Positive feedback (dashed arrow) of the idler to the amplification process. Only the first two of all the infinite local interactions are shown. Taken from [3].

Before moving on, it is worth to mention that this is of course the ideal situation, since perfectly monochromatic waves were assumed at the beginning and the *no depletion approximation* was applied. In practice, other important factors such the effective

nonlinear optical coefficient d_{eff} or the parasitic absorption⁴ of the medium have significant negative effects on the efficiency of the process. Besides, while a great length of the crystal means and exponential growth of the gain, it also implies greater losses by dispersion, specially threatening for ultrashort pulses with broad frequency ranges. But on the other hand, in Eq.(1.13) one can see that the gain scales exponentially with the square root of the pump intensity I_3 (included in Γ , in turn included in g , see Eqs.(1.10) and (1.11)). Therefore, ultrashort pulses are exceptional candidates for OPA applications due to their high peak powers. Analogously, the gain also scales exponentially with the square root of the product of the signal and idler frequencies. This suggests to use high frequencies, but conversely, this means larger differences in group velocities of the interacting pulses, which contributes negatively to the efficiency. A certain balance must therefore be accomplished for optimal performance⁵.

1.1.2 Phase-matching

The efficient propagation of various waves at different frequencies in the same medium is a relevant challenge. Bearing in mind the dependency of the refractive index with the frequency, each one of the nonlinearly generated fields will experience a different refractive index, i.e., they will propagate at a different velocity and interfere destructively in the most general case. To observe a macroscopic net effect, their phase velocities should be tuned in such a way that their phases match and their interference becomes constructive. More precisely, the speed of the corresponding forcing term $\partial^2 P_{NL}/\partial t^2$ should be equal to the phase velocity of the respective nonlinearly generated beam, so that the components generated in different points of the crystal share the same phase and experience constructive interference [3]. From Eq.(1.6), the term corresponding to the A_3 field propagates as

$$e^{i[\omega_3 t - (k_1 + k_2)z]}, \quad (1.14)$$

while the beam at ω_3 propagates as

$$e^{i(\omega_3 t - k_3 z)} \quad (1.15)$$

Thereby, their corresponding phase velocities are

$$v_{P_{NL}} = \frac{\omega_3}{k_1 + k_2} \quad \text{and} \quad v_3 = \frac{\omega_3}{k_3} \quad (1.16)$$

Forcing them to be equal, the relation between their wave numbers under the phase-matching condition is reached:

$$v_{P_{NL}} \stackrel{!}{=} v_3 \quad \Rightarrow \quad k_1 + k_2 = k_3 \quad (1.17)$$

This equation reminds to a momentum conservation relation, and it is exactly what it becomes by multiplying it by the *reduced Planck's constant*, $\hbar$:

$$\hbar k_3 = \hbar k_2 + \hbar k_1 \quad (1.18)$$

⁴In principle, the OPA process should not deposit any energy on the nonlinear crystal since it operates in its virtual states; however, in reality there is always a *parasitic* linear or multiphoton absorption, which becomes specially relevant for higher frequencies.

⁵What is usually done in practice is carrying a step-by-step amplification process using a controlled crystal length in each.

In fact, if nonlinear interactions are described as collisions of photons, the corpuscular interpretation of phase-matching is momentum conservation during such collisions [3]:

$$\hbar\omega_3 = \hbar\omega_2 + \hbar\omega_1 \quad (1.19)$$

If some of the energy is lost in the process, e.g. due to a non-ideal nonlinear coefficient d_{eff} , by a misalignment or by parasitic absorption of the crystal or any other optics, $k_3 \neq k_1 + k_2$ and neither momentum nor energy is conserved, thus efficiency gets lowered.

Eq.(1.17) holds when the pump and signal fields propagate in the same direction, but it can be easily extended to the non-collinear case, which is the most general one:

$$\vec{k}_3 = \vec{k}_2 + \vec{k}_1 \quad (1.20)$$

Non-collinear OPAs (NOPAs), like the one used in this thesis, offer two main advantages:

1. The angle between the pump and the signal adds another degree of freedom to play with in order to reach the phase-matching condition.
2. Spatial separation of the signal from the rest of the beams becomes feasible without the use of any filters or optics which could alter the beam profile.

The fundamental question which is yet to be clarified is how the phase-matching condition is really accomplished in practice. It can be easily shown that this is not possible in any isotropic medium with either positive or negative dispersion by taking Eq.(1.17) and using $k_j = \omega_j n_j / c$:

$$\begin{aligned} n_3\omega_3 &= n_2\omega_2 + n_1\omega_1 & \xRightarrow{\omega_2=\omega_3-\omega_1} & n_3\omega_3 = n_2(\omega_3 - \omega_1) + n_1\omega_1 \\ & \Rightarrow \omega_3(n_3 - n_2) = \omega_1(n_1 - n_2) \end{aligned} \quad (1.21)$$

Assuming $\omega_1 < \omega_2 < \omega_3$ and an isotropic medium with positive dispersion ($\partial n / \partial \omega > 0$), then $n_1 < n_2 < n_3$. The left-handed side of the equation will thus be positive while the right-handed one, negative, so there is no solution. Taking a medium with negative dispersion instead, ($\partial n / \partial \omega < 0$), the final conclusion remains the same.

The phase-matching condition can be reached in the first place exploiting birefringence and using uniaxial materials. For a detailed derivation, please refer to the **Appendix**. Depending on the polarization of the fields, uniaxial crystals are classified according to **Table 1.1**, where “o” stands by “ordinary” and “e” stands by “extraordinary”. Polarization states are always displayed such that the first one corresponds to ω_1 , the second to ω_2 and the third to ω_3 .

Table 1.1: Types of uniaxial crystals and possible polarization configurations for each.

	Negative ($n_o > n_z$)	Positive ($n_o < n_z$)
Type 0	ooo	eee
Type I	ooe	eeo
Type II	oeo eoe	oeo eoo

Nevertheless, not all of these configurations can be phase-matched with this method. In particular, fields must have different polarizations in order to experience different refractive indexes, so Type 0 configurations are automatically discarded⁶. For the rest of them, the ordinary component will propagate with refractive index n_o , while the extraordinary one will propagate with $n_e(\theta)$, where θ is the angle between the direction of propagation $\vec{s}$ and the optical axis. The phase-matching angle is the angle for which $n_e = n_o$, i.e. the ordinary and extraordinary components propagate at the same velocity.

For the nonlinear crystal used in the experiment, BaB_2O_4 , (typically known as BBO) phase-matching is only accomplished if ω_3 carries the extraordinary component, since it is a negative uniaxial material. Besides, NOPAs rely on another extra angle α between the pump and the signal, which makes the calculation of θ_m more elaborate. Fortunately, there are already established numerical methods for this [6].

To conclude, it is important to note that all this treatment can be adapted to ultrashort pulses. In the previous pages, three monochromatic waves, one for each field, were assumed. According to the Fourier Theorem, light pulses are just a superposition of many monochromatic waves, so in this case one will have to deal with three *packs* of many monochromatic waves (the shorter the pulse is, the more superposed monochromatic waves will be). As a result, many other important factors associated with light pulses, such as group velocities, group velocity mismatches (GVM) or group velocity dispersions (GVD), must in this case be taken into account. Although a complete derivation of the performance of OPA with light pulses would require some extra pages, a set of important tips in the design of ultrafast OPAs will be given in the next section.

1.1.3 Design criteria of ultrafast OPAs

1. Pulse-splitting length

At the end of **subsection 1.1.1**, the problem of dispersion in ultrashort pulses due to their wide spectrum was already mentioned. In order to accomplish net gain, temporal overlapping between the pump and the signal pulses is crucial. We call *pulse-splitting length* to the distance it takes for the pulses to temporally separate (or *walk-off*) due to the relative group delay accumulated during their propagation. Therefore, the pulse-splitting length is directly related to the length of the nonlinear crystal used in the process. It is important to note that it decreases with the pulse duration and for increasing group velocity mismatch. In other words: it will be always more difficult to temporally overlap shorter light pulses propagating with a greater difference in their group velocities. Given that gain also increases with crystal length, an equilibrium must be reached.

⁶Another alternative to optimize efficiency is the so-called *quasi-phase-matching* (QPM) regime, and in this case, fields can propagate with parallel polarizations. Applying a periodic modulation to the sign of the nonlinear coefficient d_{eff} , an average macroscopic net exchange of energy between fields is obtained despite a phase-matching condition not locally satisfied.

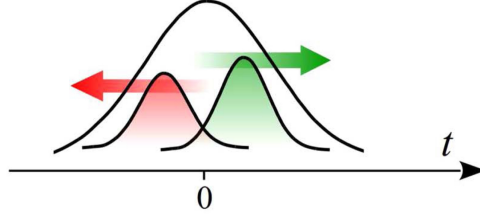


Figure 1.4: Illustration of the temporal walk-off of two light pulses. In contrast with classical optical amplifiers, OPAs do not have a energy storage capability; the gain is present only during the temporal overlap between the interacting pulses. Taken and adapted from [3].

2. Broadband phase-matching

The ideal amplifier would be that which for a fixed pump frequency provides great gains over a wide range of different signal wavelengths. However, mathematically talking it is only possible to maximize gain for a given set of frequencies. The typical procedure is to perform a first-order Taylor expansion of the phase-mismatch $\Delta k = k_3 - k_2 - k_1$ around a slightly modified signal frequency, $k(\omega_1 + \Delta\omega_1)$. By doing this, one can verify that the broadest phase-matching condition is fulfilled when the group velocity of the signal matches the projection of the group velocity of the idler along the signal direction [3]. Note how in the previous point the importance of the temporal overlapping between **pump** and **signal** was highlighted; now, the relevance of the synchronization in group velocities between the **signal** and the **idler** comes into play, evincing again the relevant role of the idler.

3. Passive CEP stabilization through the idler beam

The electric field of an ultrashort laser pulse can be described as a fast sinusoidal oscillation of angular frequency ω_0 (*carrier wave*) travelling at phase velocity $v_\varphi = \omega/k$, multiplied by a more slowly varying *envelope* $A(t)$ travelling at group velocity $v_g = d\omega/dk$:

$$E(t) = A(t) \cos(\omega_0 t + \varphi_{CEP}) \quad (1.22)$$

φ_{CEP} is the so-called *carrier-envelope phase* (CEP), and it denotes the relative phase of the carrier with respect to the envelope maximum, which for convenience it is assumed to be at $t = 0$ (see **Figure 1.5**). Femtosecond laser sources are mode-locked, and here a pulse train is usually generated from a single pulse circulating in the laser resonator. Along the propagation of the pulse in the cavity, the group and phase velocities will be different due to dispersion effects in the crystal and some additional contributions from the mirrors and intracavity air [7]. If one assumes that the pulse propagates in the z direction, the CEP shift at position z can be described by [8]

$$\Delta\varphi_{CEP}(z) = \omega_0 \left(\frac{1}{v_g} - \frac{1}{v_\varphi} \right) z = \frac{\omega_0^2}{c} \frac{dn(\omega)}{d\omega} \Big|_{\omega_0} z, \quad (1.23)$$

where ω_0 is the angular frequency of the incident field, $n(\omega)$ is the refractive index of the dispersive medium and c is the velocity of light.

Its stabilization implies the generation of light pulses with a reproducible electric field profile. While this is not a prerequisite for the generation of attosecond pulse trains [8], it is for the generation of single isolated attosecond pulses. Another of the most

important applications of the idler beam is the stabilization of the CEP. By linking the CEPs of the pump, signal and idler, one can see that OPAs produce a CEP stable idler when the seed (the weak pulse at the signal frequency which starts the OPA process, hence its name) is generated with the same pulse used as the pump. Comparing the CEP of the signal with the one of the idler, it is therefore possible to stabilize the former. This technique is called *passive CEP stabilization* [3, 7, 9].

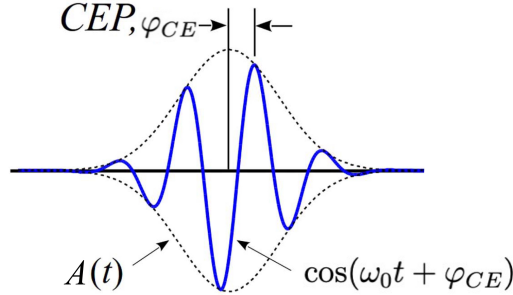


Figure 1.5: Carrier-Envelope Phase. Taken and adapted from [3].

4. Pump-pulse parameters

- **Wavelength:** in order to avoid energy losses and accomplish optimal efficiencies, the pump, signal and idler frequencies must be included in the transparency range of the nonlinear crystal(s) used. As commented above, the gain scales with the frequencies of the pump and signal pulses, but that implies larger temporal walk-offs due to their greater GVM and therefore shorter pulse-splitting lengths.
- **Energy:** the first thing which must be regarded is the damage threshold of the optics in use. Additionally, very high peak intensities may start spurious and undesired second and third-order nonlinear processes with a negative impact in the efficiency. According to [3], for BBO the pump pulse energy and spot-size should be chosen to get a pump peak intensity of the order of $100\text{-}150 \text{ GW cm}^{-2}$, with a clean beam profile and avoiding hot spots. Pulse-front tilting techniques to the pump could be convenient to avoid spatial chirp of the amplified pulse.
- **Duration:** OPAs can generate pulses significantly shorter than the pump laser. The most important thing regarding this issue is that the duration of the seed must be shorter or comparable to the pump pulse to ensure their temporal overlap (see **Figure 1.4**).

5. About the crystal

Apart from transparency and damage threshold, the nonlinear coefficient d_{eff} plays a vital role, too, so high nonlinear crystals with low refractive indexes are preferred in order to optimize it. Besides, the amplification bandwidth must be taken into account, as well as the suitable type of crystal (see **Table 1.1**). Last but not least, the crystal length must be correctly chosen so that the total gain is greater than dispersion losses, as previously discussed.

6. Pulse compression

This is an optional stage of an OPA, which is usually carried in sub-50 fs light pulses

in order to compensate the spectral phase and chirps accumulated by the amplified pulses, thereby accomplishing the time limited (TL) pulse duration given by the Fourier transform. Various techniques are available for this task, such as Brewster-cut prisms, grating pairs, chirped mirrors, pulse shapers or acousto-optic programmable dispersive filters. Nevertheless, the introduction of new optics in the system implies the introduction of new energy losses (e.g. by absorption), so pulse compressions should be carefully studied before their installation and applied only when strictly needed.

7. Repetition rate

Depending on their application, OPAs can operate at many different powers and repetition rates. However, at very high values of these parameters, thermal effects like the mentioned *parasitic* absorption of the nonlinear crystal may become relevant, leading to temperature gradients which are in turn responsible of beam distortions or deviations from the phase-matching conditions.

1.2 White Light Generation

One of the main purposes of using OPAs is the possibility of amplifying a wide range of different wavelengths by a customized tuning of the phase-matching condition. Nevertheless, it should be noted from **Figures 1.1** and **1.2** that in order to amplify a certain frequency, it must be sent to the OPA along with the pump. Therefore, the great advantage that OPAs offer becomes in practice completely restrained if the signal is frequency-fixed, given that despite being able to amplify a wide range of different values, only one signal will in the end participate in the process. This is why *white-light* is an excellent seed for OPAs: it provides a broad spectrum spanning from the visible to the infrared from which the desired signal can be freely chosen, and using phase-matching, amplified.

White-light generation (WLG) is a third-order nonlinear process. Taking a look back to Eq.(1.2), attention is now shifted to the third-order nonlinear term, $P^{(3)}(t) = \varepsilon_0 \chi^{(3)} E^3(t)$. Before understanding the process of WLG, the Optical Kerr Effect should be addressed.

1.2.1 Optical Kerr Effect

Let $\Delta\chi$ be the change in susceptibility due to the $\chi^{(3)}$ term. Bearing in mind that

$$n = \sqrt{1 + \chi}, \quad (1.24)$$

the refractive index can therefore be expressed in terms of this change in susceptibility as follows:

$$n = \sqrt{(1 + \chi) + \Delta\chi} = n_0 \left(1 + \frac{\Delta\chi}{n_0^2} \right)^{1/2} \quad (1.25)$$

where n_0 is the refractive index of the medium that contributes to the linear part of the susceptibility. Assuming a small nonlinear term compared to the linear one, the above equation can be expanded using the Taylor series, such that [4]:

$$n = n_0 + \frac{\Delta\chi}{2n_0} \quad (1.26)$$

Rather than its true value, what is most important about $\Delta\chi$ is its dependency, and in order to obtain it, the third-order polarization term needs to be explicitly calculated. Assuming three incident fields, all of them oscillating at the same frequency for the sake of simplicity, $E(t) = E_0\cos(\omega t)$, thus

$$\begin{aligned} P^{(3)}(t) &= \varepsilon_0\chi^{(3)}[E_0^3\cos^3(\omega t)] = \frac{\varepsilon_0\chi^{(3)}E_0^2}{4} [E_0\cos^3(\omega t) + 3E_0\cos(\omega t)] = \\ &= \left(\frac{\varepsilon_0\chi^{(3)}I_0}{4}\right) E_0\cos^3(\omega t) + \underbrace{\left(\frac{3\varepsilon_0\chi^{(3)}I_0}{4}\right) E_0\cos(\omega t)}_{\Delta\chi}, \end{aligned} \quad (1.27)$$

where $I_0 = E_0^2\cos^2(\omega t)$ is the intensity of the electric field. While the first term in the above equation corresponds to the third-harmonic generation (THG) process, the parenthesis in the second term represents the change in susceptibility, $\Delta\chi$, and as it can be observed, it depends on the intensity of the electric field. As a consequence of Eq.(1.26), this thereby means that the refractive index must also depend on the intensity of the electric field *at high enough intensities*, where the $\chi^{(3)}$ term has a considerable influence and the change in susceptibility $\Delta\chi$ becomes relevant⁷. Rewriting,

$$n(I) = n_0 + n_2(I)I, \quad (1.28)$$

where $n_2 = 3\varepsilon_0\chi^{(3)}/8n_0$ is the nonlinear refractive index, which describes the temporal birefringence induced in a certain Kerr medium due to the disturbance of the atomic motions from equilibrium and the distortion of the electron clouds caused by an intense incoming beam [10]. The quadratic dependence of the refractive index on the incident optical field is the basis of the Kerr Effect, and leads to various nonlinear terms which interact and trigger WLG.

Transverse Optical Kerr Effect: Self-focusing effect

A direct consequence of Eq.(1.28) is that the refractive index becomes larger in areas where the intensity is greater according to the spatial profile of the incoming beam. Therefore, the medium experiences an intensity pattern in its *transverse* plane and thereby acts a graded-index material [4]. Assuming for example a Gaussian-shaped laser pulse and $n_2 > 0$, the refractive index will then be maximized at the central part, while the outer one will in contrast experience a lower value, i.e. a lower phase velocity which in turn curves the phase front. In other words, the pulse experiences *self-focusing* (when $n_2 < 0$, the effect is the opposite):

The self-focusing effect takes place when the optical power is greater than

$$P_{crit} = \frac{\pi(0.61)^2\lambda_0^2}{8n_0n_2}, \quad (1.29)$$

where λ_0 is the wavelength of the laser source in vacuum. This threshold power takes into account the natural divergence of the beam due to diffraction effects. If $P \gg P_{crit}$, the beam is expected to collapse and breakup in many components each carrying approximately power P_{crit} [5]. However, at moderate optical powers there are other defocusing effects such as multi-photon absorption from the valence to the conduction

⁷Of course, assuming that the material exhibits a non-zero $\chi^{(3)}$ term, i.e., assuming *Kerr media* [4].

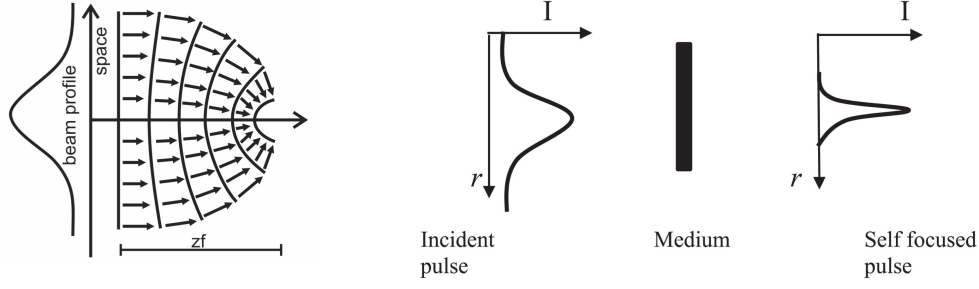


Figure 1.6: Self-focusing effect, also known as *Kerr lens*, for a Gaussian input beam. Adapted from [4] and [10].

band, saturation of the Kerr effect, nonlinear absorption, etc, which prevent this situation. In this experiment, a solid sapphire (Al_2O_3) plate was used in order to create white light, and in these cases, the most important influences come from the GVD and plasma defocusing [10]. The balance between self-focusing and defocusing mechanisms is called *filamentation* or *self-trapping of light*, and arises when the optical power becomes exactly equal to P_{crit} .

Longitudinal Optical Kerr Effect: Self-phase modulation (SPM)

The longitudinal Optical Kerr Effect is the main responsible of the spectral broadening and thus WLG. The angular frequency ω of a wave is related to the rate of change of its phase $d\phi/dt$, and in turn, the phase is related to the refractive index.

Expressing the total phase as the sum of a linear and a nonlinear contribution:

$$\phi_{tot} = kL \cong \left(\frac{\omega_0}{c} nL \right) = \frac{\omega_0}{c} n_0 L + \frac{\omega_0}{c} n_2(I) L = \phi_l + \phi_{nl}, \quad (1.30)$$

where L is in this case the length of the sapphire plate responsible for the effect, ω_0 is the frequency of the incident laser beam.

Taking now into account the dependence of the refractive index with the frequency, Eq.(1.28) can be rewritten as

$$n(I, \omega) = n_o(\omega) + n_2(I, \omega)I, \quad (1.31)$$

which allows to express the frequency change $\Delta\omega$ due to the nonlinearity as

$$\Delta\omega = -\frac{d\phi_{nl}}{dt} = -\left(\frac{\omega_0}{c} \right) L \frac{dn_2(I, \omega)}{dt} \quad (1.32)$$

This is the so-called *self-phase modulation*, and its effect is shown in **Figure 1.7**. Its physical interpretation is that at the beginning of the laser pulse, $dn_2(I, \omega)/dt$ is a positive quantity. From Eq.(1.32), the frequency change $\Delta\omega$ turns more negative, i.e. the phase front becomes delayed and this results in a reduction of the frequency of the incident pulse. Analogously for the trailing part, $dn_2(I, \omega)/dt$ turns negative and frequency gets increased. Consequently, the pulse gets *chirped* and its spectrum, broadened. It is important to note how for ultrashort pulses (with great peak intensities) and for great enough values of L , the previously discussed self-focusing effect (also known as *spatial self-phase modulation*), which is also acting meanwhile, strengthens the SPM by increasing the intensity inside the medium, i.e. contributing to the creation of new frequencies.

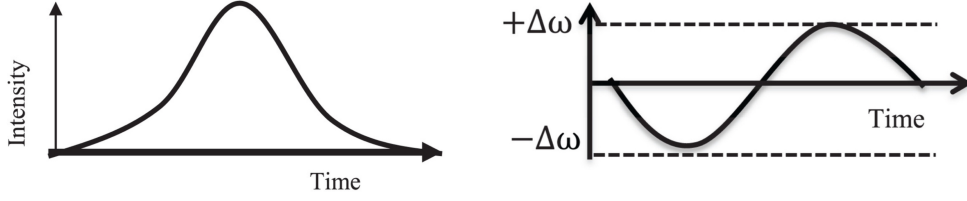


Figure 1.7: Spectral broadening ($\omega_0 \pm \Delta\omega$) due to SPM for a Gaussian beam. Adapted from [4].

Self-steepening

This is another nonlinear consequence of the Kerr Effect, which appears instantaneously during filamentation. As commented before, the main mechanism avoiding a total collapse of the beam due to self-focusing in solids is the influence of the GVD, which accounts for the differences in group velocity among the new generated frequencies. From the following equation [10]:

$$v_g = \frac{c}{n - \lambda \frac{\partial n}{\partial \lambda}} = \frac{c}{(n_0 + n_2(I)I) - \lambda \frac{\partial(n_0 + n_2(I)I)}{\partial \lambda}} \quad (1.33)$$

it can be seen that it does not only depend on the wavelength of the incoming beam, but also on its intensity (as well as on the variations of linear refractive index ($\partial n_0/\partial \lambda$), nonlinear refractive index ($\partial n_2/\partial \lambda$) and spectrum ($\partial I/\partial \lambda$)). Regarding again a Gaussian beam as in previous examples, the central part will in this case experience a greater intensity than the wings of the pulse, and therefore, will propagate with a lower group velocity. This leads to a shift of the peak of the pulse towards the trailing edge, forming a steep slope, hence its name. Self-steepening can be used for the generation of few cycle pulses since it dramatically shortens the pulse duration, even in solids. If the effect is such that the intensity changes instantaneously and the steep becomes infinite, the formation of an optical shock wave takes place.

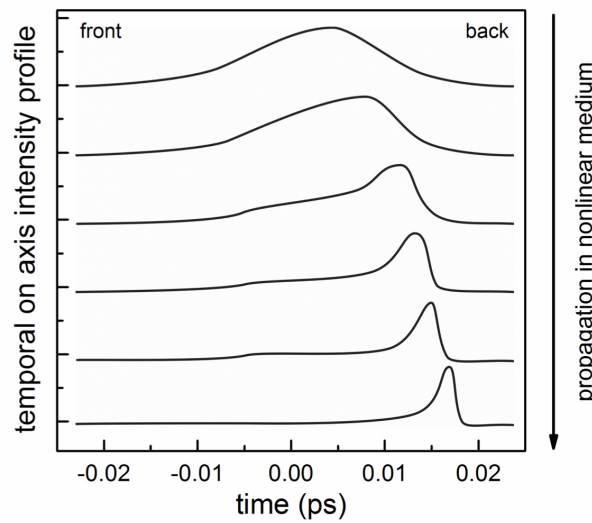


Figure 1.8: Evolution of the temporal intensity profile of an initial Gaussian pulse due to self-steepening. Taken and adapted from [10].

Practical generation of white light

First observed in 1970 by focusing intense picosecond pulses into glass, WLG is still a subject of research nowadays since it can be generated through many diverse ways involving gases, liquids and solids. Each kind of process leads to different white-light beam properties which need to be optimized according to its purpose. As commented before, a sapphire plate irradiated with femtosecond laser pulses in the near infrared (NIR) was used in this experiment.

When it comes to solids, the bandgap of the medium must be carefully regarded in the first place, since it determines the short wavelength continuum cut-off and the necessary pump energy [10]. According to [11], WLG generation is not possible below a threshold bandgap of 4.7 eV. Additionally, the threshold power for continuum generation coincides with the critical power for self-focusing, shown in Eq.(1.29). Above this value, the width of the continuum increases with the bandgap of the medium [10,11]. Due to self-focusing (counterbalanced by GVD), the intensity of the beam increases at certain parts, which in turn brings into play both SPM and self-steepening. A broad spectrum ranging from the visible to the infrared is then generated, which projected into a screen, shows a bright, white spot in the central part surrounded by a concentric, rainbow-like pattern. This white disk, commonly referred as the *supercontinuum*, contains only 10% of the energy [10] and can be easily separated from the rest of the beam by means of an iris properly positioned.

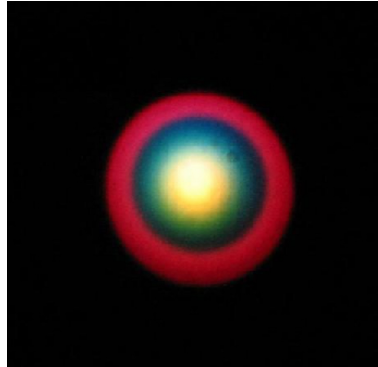


Figure 1.9: Example of supercontinuum. Taken and adapted from [10].

In combination with Ti:Sa femtosecond pulses, sapphire (with a bandgap of 7.5 eV) is an ideal nonlinear crystal candidate due to the modest pump energies it requires and the excellent properties it brings to the generated white light: high-quality beam profile and low pulse-to-pulse fluctuations. Both intensity and stability are better in comparison with, for example, fused silica glass [11]. On the other hand, it is true that due to self-steepening pulses become a little asymmetrical.

Other parameters influencing the efficiency of WLG are the focusing geometry, and of course, the crystal length. As explained before it strengthens SPM and self-steepening, the main supercontinuum mechanisms, but at the same time it favors the undesired chromatic and temporal dispersion of the new wavelengths [10]. Thus, a balance must again be reached.

Apart from the previously described third-order effects, other parametric processes such as stimulated Raman scattering (SRS), four-photon generation, cross-phase modulation

(XPM), cascading, light up- and downconversion also participate in WLG. Although some literature sources highlight the effects of some of them over the others (specially SRS and four-photon generation), they all agree in the fact that the main triggers are the three mechanisms described above.

1.3 Velocity Map Imaging

In order to realize how matter behaves a deep inspection of its internal structure is required. Systematic studies about particle identification were initiated after Thomson's discovery of the electron in 1897. By varying the intensity of two crossed electric and magnetic fields, he was able to determine not only their velocity, but also their charge-to-mass ratio [12]. This was the first mass-spectrograph, and their performance improved significantly during the first half of the 20th century.

The possibility of identifying different particles under the same experimental conditions came with the invention of Time Of Flight mass spectrometers (TOF-MS) in the decade of 1940. In these devices, particles are ionized in the region between two electrodes (called *repeller* and *ground*) held at a certain potential difference. Due to the resulting electric field, ions get accelerated in one direction and are directed to a field-free ("drift") region after passing through the hole of the corresponding electrode. Measuring the time it takes for them to *fly* and get detected, their masses can be calculated. TOF-MS laid the foundation of velocity map imaging (VMI) spectrometers.

The next step in the characterization of reaction products was the determination of their kinetic energies and angular distributions, which reflect intimate details of the potential energy surfaces on which the dynamics take place [13]. While the main method for the former calculation was brought by the Photo-Fragment Translational Spectroscopy (PFTS), still in use today, the latter was possible by the development of the Photo-Fragment Imaging (PFI) technique, reported by Chandler and Houston in 1987 [14]. Combining a position-sensitive ion detector and a Charged-Coupled Device (CCD), they found a way to project the three-dimensional spatial distribution of *photofragments* (the so-called *Newton Sphere* (NS)) onto the two-dimensional surface of the detector [15]. It was soon adopted for a wide range of photodissociation studies, but it had a severe drawback regarding its resolution, as they themselves stated in their paper:

*"Presently, the main limitation to the spatial resolution of the projection
[of the 3D NS onto the 2D surface of the detector]
is due to the fact that the molecular beam width is significant on the scale of the
image."*

This is when André T. J. B. Eppink and David H. Parker took over and developed the Velocity Map Imaging (VMI) technique in 1997 [16], which did not only solve that problem, but it managed to measure both kinetic energies and angular distributions at the same time.

1.3.1 Traditional setup and working principle

The original configuration of the VMI spectrometer of Eppink and Parker is shown below:

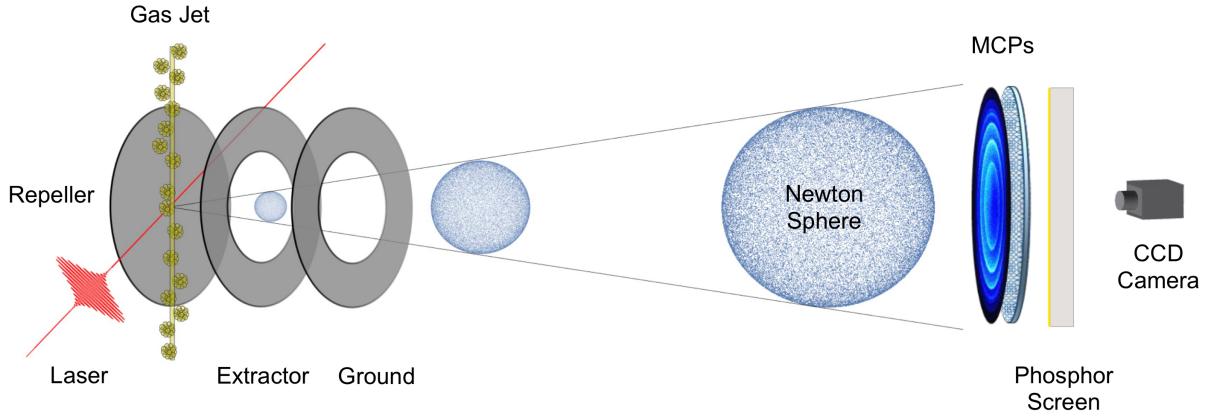


Figure 1.10: Eppink-Parker-Design of the VMI spectrometer. Adapted from [17,18].

The principal elements of the setup are three circular electrodes, *repeller*, *extractor* and *ground*; a flight tube and a detection system made up of a pair of *microchannel plates* (MCPs), a phosphor screen and a CCD camera⁸.

Its main purpose is to investigate the velocity-distribution of charged particles obtained through photoionization as a result of the interaction of high-energy photons with a molecular or atomic beam. In the most general case, the resulting fragments in these collision processes scatter isotropically and expand as a series of concentric Newton Spheres. Their characterization is key in order to fully understand the dynamics governing the process. In the setup, the interaction point between the laser pulse and the molecular beam is located between the repeller and the extractor. Keeping in mind the charge sign of the photofragments under study, an appropriate potential difference is set between the latter. As a consequence, an electric field arises and photofragments are accelerated towards the flight tube. After passing through both of the holey electrodes and because of the ground being (typically) kept at 0 V, they enter into a field-free region where they keep propagating until they get finally projected into the first component of the detection system: the microchannel plates. Essentially, they are an array of pores, each of them working as an electron multiplier tube. Between its front and backside, a voltage is applied for two reasons: the first of them is allowing its internal coating to release a great amount of electrons when a high-energy photofragment manages to go through; and the second, to guide the newly generated electrons towards the end of the MCP. The subsequent collisions of all these new electrons with the walls of the pore leads to an *electron avalanche* which amplifies the signal. Usually, another MCP is placed right next to the first one in a double-chevron configuration to enhance collisions and get an even greater amplification⁹, leading to final net gains typically in the order of 10^6 . **Figure 1.11** illustrates this mechanism. Following the exit of the second MCP, electrons are directed towards a phosphor screen, which after absorbing their kinetic energies emits light at the point where they collided. Finally,

⁸Another popular detection system are delay-line anodes.

⁹The VMI spectrometer used in this thesis has only one MCP.

the CCD camera records the fluorescent radiation and sends it to the PC where data is later analysed.

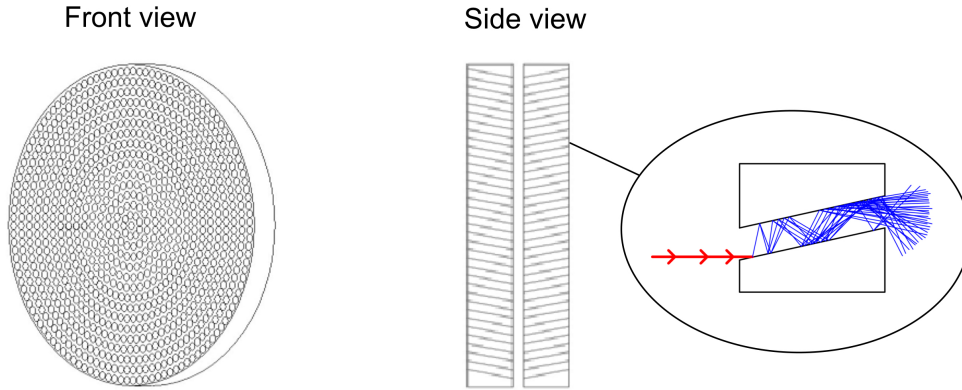


Figure 1.11: Front and side views of the pair of microchannel plates in double-chevron configuration. Adapted from [14].

It is important to mention that VMI spectrometers are often shielded with a μ -metal coating in order to prevent the influence of external magnetic fields (such as the Earth’s magnetic field) on the photofragments’ dynamics inside the flight tube. In order to avoid any other kind of perturbation, the spectrometer is kept in vacuum.

The origin of the problem regarding the resolution is that the interaction point is not really a point source, but has a finite volume. In the original experiments, the repeller and extractor plates consisted of a pair of grids that provided a homogeneous electric field, ensuring a direct spatial mapping of the photofragments onto the detector [15]. Nevertheless, this would only work if the source of photofragments is pointlike, which is not the case considering the finite size of the laser and molecular beam. As a consequence, photofragments with the same velocity but different initial points of origin got projected on non-identical locations of the detector surface, leading to blurry images. Minimizing the interaction volume reduced the blurring, but it naturally led to a reduction of signal strength. What Eppink and Parker did was removing the grids in the center of the electrodes in order to create an inhomogeneous electric field able to focus the Newton Sphere on the detector. In other words, they made electrodes work as electrostatic lenses which under the correct voltage ratio compensated the effect of the finite size of the interaction region. Consequently, photofragments with the same initial velocity arrived at the same position on the detector regardless of where they were produced, hence the term “velocity mapping” [14]. The result is an image containing information of both the velocity and angular distributions: while a greater modulus of the photofragments’ velocity vectors corresponds to greater radii of the concentric circumferences, their different directions explain the angular distribution¹⁰. Additionally, with the omission of the mesh grid they increased the fraction of photofragments that reached the detector, which also contributed positively to the resolution of the device [19]. **Figure 1.12** shows the improvement in resolution of this method if compared to the photo-fragment imaging technique:

¹⁰The polarization of the incident beam plays an important role here. See next section.

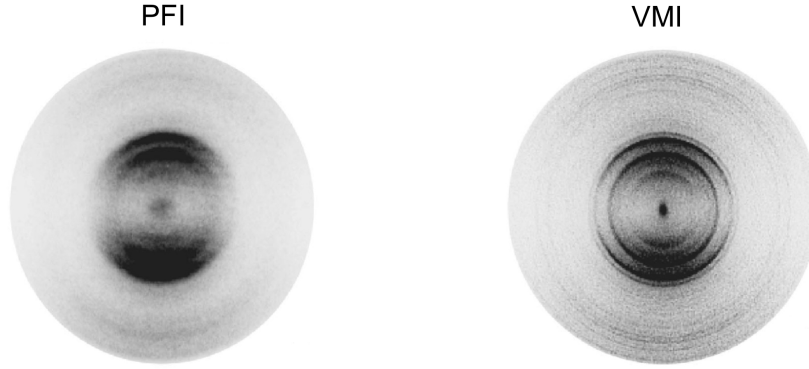


Figure 1.12: Comparison between the velocity and angular distribution images of oxygen atoms from the dissociation of O_2 using PFI and VMI. Taken from [16].

The second great advantage of the Eppink-Parker-Design is that depending on the ratio between repeller and extractor voltages, not only the velocity distribution can be imaged (VMI), but also the spatial one (SMI) [19, 20].

To conclude with this section, it is worth to mention that this device measures specifically the velocity of the photofragments, not their kinetic energies. Since the kinetic energy is quadratic in velocity, $E = 1/2mv^2$, the experimental consequence is that a peak at low kinetic energy will be narrower (will have better resolution) than the same peak at high kinetic energy [18]. Since VMI is becoming increasingly popular when combined with synchrotron radiation, free-electron lasers and laser-based sources of extreme ultraviolet radiation (XUV), new configurations are nowadays being explored in order to optimize both their resolution and energy acceptances depending on the particular purpose. Some possibilities in order to achieve this are increasing the size of the detector, reducing the flight distance (so that the NS does not get expanded so much), increasing the acceleration voltages (so that the NS reaches the detector faster and does not get expanded so much) or adding electrostatic lenses to improve the focusing performance [21].

1.3.2 Image Reconstruction

As seen from **Figure 1.10**, in its conventional form the surface of the detection system is two-dimensional. Therefore, what is really measured with the VMI spectrometer is the two-dimensional projection of the three-dimensional Newton Sphere, also known as *Newton pancake*. While its size is a direct reflection of the speed of fragments, its shape is associated with their angular distribution. However, in order to physically characterize the dynamic of the process it is necessary to recover the original three-dimensional NS from the measured data, a puzzle under study since 1968. While some scientists used various reconstruction techniques with the purpose of creating a 3D density map of the tail of a bacteriophage from electron microscope images, engineers used them in order to analyse plasmas and flames from jet engines [14]. Regarding photoionization processes, it was later realized that in practice, only the central slice of the three-dimensional NS is enough to retrieve all the essential and available physical information [14, 15].

Assuming a cylindrically symmetric photofragment distribution¹¹, which can be accomplished irradiating the molecular beam with a linearly polarized laser beam perpendicular to the spectrometer axis (i.e. parallel to the detector surface), image reconstruction can be performed by means of a numerical resolution of the inverse Abel transformation. Let $f(r, y)$ be the original, cylindrically symmetric (3D) distribution along the y -axis, and $F(x, y)$ its projection (2D), according to the sketch shown in **Figure 1.13**.

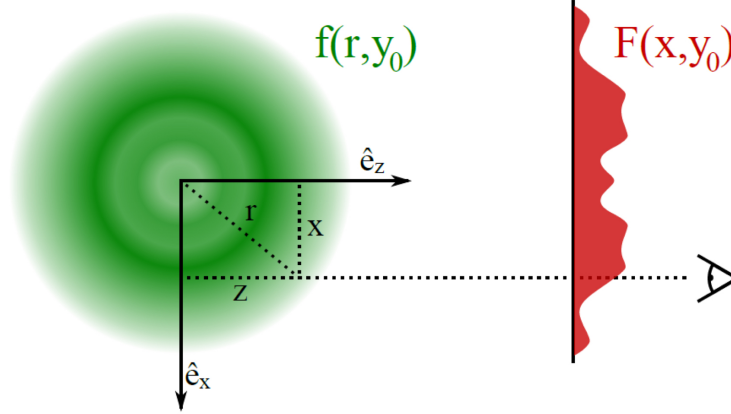


Figure 1.13: Plots of the cuts through the original 3D distribution $f(r, y)$ and the projected one $F(x, y)$, for a fixed $y = y_0$. Taken from [20].

From the perspective of an infinitely far observer looking along the z -axis:

$$F(x, y) = \int_{-\infty}^{+\infty} f(r, y) dz = 2 \int_0^{+\infty} f(r, y) dz \quad (1.34)$$

Using cylindrical coordinates, the distance to the y -axis r contains information of x and z :

$$r^2 = x^2 + z^2 \quad \Rightarrow \quad dz = \frac{r}{\sqrt{r^2 - x^2}} dr, \quad (1.35)$$

and replacing in Eq. (1.34):

$$F(x, y) = 2 \int_{|x|}^{+\infty} \frac{r f(r, y)}{\sqrt{r^2 - x^2}} dr \quad (1.36)$$

This is the so-called Abel-transform, and it shows how to calculate the 2D projection of a 3D distribution. Note however that in this case what is known is $F(x, y)$. It can be shown that [20, 22]:

$$f(r, y) = -\frac{1}{\pi} \int_{|r|}^{+\infty} \frac{dF(x, y)}{dx} \frac{1}{\sqrt{x^2 - r^2}} dx, \quad (1.37)$$

which is the *inverse Abel transform*. Nevertheless, its analytical solution is impractical due to the singularity at the origin $x = r$, the extreme sensitivity to noise and the

¹¹In principle, random gas-phase collisions create Newton Spheres with homogeneous (isotropic) surface patterns, thus anisotropic surface patterns are the result of a selected directionality in the process. Apart from linear polarization of the irradiating beam, one can also accomplish preferred directions crossing two molecular beams at a certain angle, selecting special molecular and angular momenta for the reactants or changing the molecular axis alignment [15].

finite number of pixels of the CCD camera (responsible of $F(x, y)$ being not continuously differentiable). Instead, several numerical approaches have been developed, the use of which depends on the specific case under study. Some examples are the Basis Set Expansion (BASEX) -Method [23], which uses a certain basis of functions $\bar{f}_k$ in the space of distributions f for which the projections $\bar{F}_k$ are well known and analytically integrable [20]; the pBASEX-Method [24] (used in this thesis), which includes cylindrical coordinates and therefore depends on less parameters; the MEVELER-Method [25] or the Vrakking's algorithm [26]. The Onion-Peeling technique [27] and some filtered backprojection methods are also available [15].

If on the other hand the photofragment distribution does not possess a cylindrical symmetry, forward convolution techniques become more suitable. These methods try either to reconstruct the whole 3D Newton Sphere, or to sample only its central 2D slice. Some of the most popular examples of *slice imaging* are Optical Slicing, Non-Optical Slicing and DC-Slicing [14].

1.4 Reconstruction of Attosecond Beating By Interference of Two-photon Transitions

Significant improvements in ultrafast laser technology during the last twenty years have given rise to completely new and fascinating fields of research. Just as femtosecond (10^{-15}s) pulses allowed the resolution of molecular movements, even shorter light pulses in the attosecond (10^{-18}s) scale promise to be ideal tools in order to resolve electron dynamics [1]. However, it is not enough to know how to create them; scrupulous characterization of the generated attosecond pulses becomes key in order to quantify electrons' behaviour. For this purpose, Reconstruction of Attosecond Beating By Interference of Two-photon Transitions (RABBITT) represents a powerful technique with applications not only in atomic and molecular physics, but also in condensed matter systems [28]. Although it was originally conceived for measuring the average duration of attosecond pulse trains, it has now shifted its main employment to a variety of photoionization experiments where attosecond temporal resolution is desired.

1.4.1 Motivation

Bearing in mind Heisenberg's Uncertainty Principle

$$\Delta E \cdot \Delta t \geq \frac{\hbar}{2} \quad (1.38)$$

and Planck's energy formula

$$E = h\nu = \frac{hc}{\lambda}, \quad (1.39)$$

it becomes clear how for *small* Δt , ΔE must be *large* enough to fulfill the inequality. In other words, very large bandwidths are required to support attosecond pulses, typically covering the optical spectrum from the near infrared (NIR) to the extreme ultraviolet (XUV). There are currently two known kinds of sources which have demonstrated the creation of coherent radiation¹² over such bandwidths: high harmonic generation

¹²As it will be shown ahead, a fixed phase relation between pulses is the main *sin qua non* of RABBITT.

(HHG) [1, 29] and free-electron lasers [30, 31], which show a tendency to produce a train of closely spaced pulses rather than a single pulse. In this thesis, an intense laser pulse with fundamental frequency ω on the NIR was focused on a noble gas jet with inversion symmetry properties, unleashing a highly nonlinear electronic response. The resulting harmonic spectrum is thereby made up of a finite number of odd harmonic peaks separated by twice the energy of the driving field. Extending far into the XUV, this frequency comb results in a train of attosecond pulses. If each harmonic is approximated to a monochromatic wave, with infinite temporal width, the temporal intensity profile $I_X(t)$ of the pulse train can be expressed as

$$I_X(t) = \left| \sum_N A_n e^{i[\omega_n t + \phi_n]} \right|^2, \quad (1.40)$$

where A_n is the spectral amplitude of the n th harmonic, ϕ_n is its phase and N is the total number of harmonics in the pulse train [8]. By looking at this equation, one can see that the attosecond pulse train is therefore uniquely determined by its amplitude and the phase of its components. While the former is obtained from the spectrum, the latter is usually harder to quantify given that beating between harmonics occurs in subfemtosecond time scales and there exists no electronic device with that resolution. This is where RABBITT takes over.

Theoretical and experimental results for the temporal characteristics of the HHG process predict that the harmonics are locked in phase by the dynamics of the electronic wavepacket in the continuum [1]. Additionally, both numerical results and the semi-classical description of the high harmonic generation process [8] show that consecutive harmonics do not have exactly the same phase because they are emitted at different recombination times, t_{rec} , and from Fourier optics it is known that a delay in the time domain corresponds to a phase shift in the frequency domain. The relative phase between two adjacent harmonics is thereby not negligible (and this is in fact what RABBITT measures). In view of the above and assuming constructive interference between a group of neighboring harmonics, one could expect several intensity spikes per optical half-cycle of the driving laser (because of the constant frequency spacing of the harmonics, twice the fundamental frequency, 2ω). However, due to the change in spatial and temporal coherence properties when the harmonic radiation propagates through the medium, only one of the interference spikes survives in the end, forming a train of attosecond pulses spaced by half the period of the fundamental [1].

1.4.2 Description

As mentioned above, one way to obtain attosecond pulse trains is by HHG. Considering *high enough* harmonic orders is then equivalent to consider *high enough* harmonic energies, which in turn opens the possibility to ionize a certain gas if the former is greater than the ionization potential of the latter, I_p . In this case, high-energy photons pull apart valence electrons from the gas target, emitted with a kinetic energy given by Einstein's formula for the photoelectric effect:

$$E_{kin} = E_{photon} - \phi, \quad (1.41)$$

where the work function ϕ is in this case I_p and $E_{photon} = n\hbar\omega$ is the energy of the corresponding photon associated to harmonic n . The higher the considered harmonic,

the greater the kinetic energy of the emitted *photoelectron*. If these energies are now measured and plotted as a function of the intensity of the signal, the photoelectron spectrum (PES) is obtained. The PES closely resembles the spectrum of the created harmonics, where their spacing ($2\hbar\omega$) and their relative intensity are directly observed, as illustrated in **Figure 1.14**.

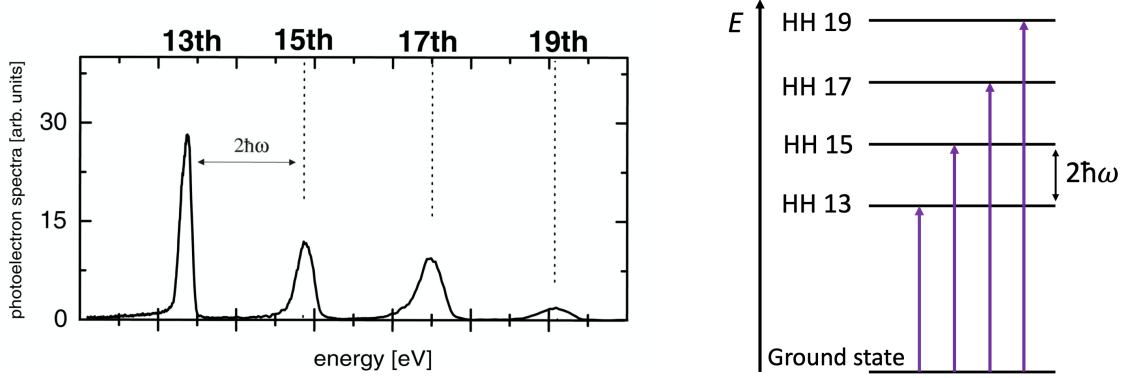


Figure 1.14: Left: example of PES adapted from [1]. Right: corresponding energy level diagram.

This is, in its most primitive format, the qualitative description of the photoemission process in the one color-field experiment (XUV only). If the original NIR field used to create the harmonic comb in the first gas target is combined with the resulting XUV radiation in the second one, photoelectrons pulled apart from the atomic gas interact with the now present NIR photons with fundamental frequency ω , undergoing one of two possible processes: either they absorb a certain number q of NIR photons, increasing their energy by $q\hbar\omega$; or they stimulatedly emit a certain number q of NIR photons, decreasing their energy by $q\hbar\omega$. Since the odd XUV harmonics are separated by $2\hbar\omega$ due to the inversion symmetry properties of the gas jet used in the HHG process, this is translated in the appearance of additional *sidebands* corresponding to even multiples of the NIR photon energy and thus located halfway between the main photoelectron peaks caused by the harmonics themselves, as shown in **Figure 1.15**.

Due to the natural poor HHG efficiency for being a highly nonlinear effect, the intensities of the individual harmonics are too weak to cause nonlinear effects on the target atoms [1]. If the NIR intensity is kept low so that no multiphoton transitions in the continuum become feasible, i.e. if only single-photon processes are considered ($q = 1$), one can safely assume that:

1. the XUV photons are the only ones responsible of ionization of photoelectrons,
2. only the two nearest harmonics contribute to each sideband peak; i.e. there are only two possibilities (two quantum paths) to reach the same final state. For example, sideband 16 in **Figure 1.15** can be reached by the absorption of one NIR photon after photoionization by harmonic 15, or by the stimulated emission of one NIR photon after photoionization by harmonic 17.
3. second-order perturbation theory is suitable for the quantum mechanical description of the problem.

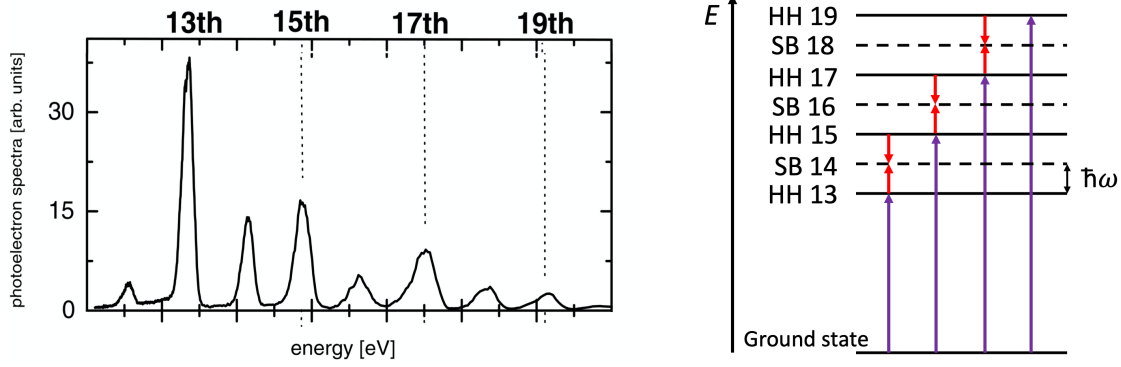


Figure 1.15: Left: example of PES adapted from [1], where the NIR field has been added to the photoionization process. Right: corresponding energy level diagram.

Due to the interference of the two possible quantum paths and also bearing in mind the link between consecutive harmonics' phases, the intensity of the sidebands can be intentionally made to oscillate by delaying the XUV and NIR fields by a time τ :

$$\begin{aligned} I_{SB}(q) &= \left| \mathcal{M}_{q-1}^{(+)} + \mathcal{M}_{q+1}^{(-)} \right|^2 \\ &= \left| \mathcal{M}_{q-1}^{(+)} \right|^2 + \left| \mathcal{M}_{q+1}^{(-)} \right|^2 + 2 \left| \mathcal{M}_{q-1}^{(+)} \right| \left| \mathcal{M}_{q+1}^{(-)} \right| \cos(2\omega\tau - \Delta\phi), \end{aligned} \quad (1.42)$$

where $\mathcal{M}_{q\mp 1}^{\pm}$ are the two-photon transition amplitudes corresponding to absorption of harmonic $q \mp 1$ and subsequent absorption (+) or emission (-) of a NIR photon [32]. They are given by Fermi's Golden Rule, which provides a way to calculate the total transition probability from an initial eigenstate Ψ_i to a continuum of eigenstates Ψ_f due to a perturbation. To first order approximation, this transition rate is given by

$$w \propto \frac{2\pi}{\hbar} |\langle \Psi_f | H' | \Psi_i \rangle|^2 = \frac{2\pi}{\hbar} |\mathcal{M}_{f,i}|^2 \quad (1.43)$$

In RABBITT, the initial eigenstate Ψ_i is taken to be the ground state, with

$$E = E_0 \quad \text{and} \quad H_0 = \frac{p^2}{2m_e} + V(\vec{r}), \quad (1.44)$$

where e is the charge of the electron, m_e is its mass, $V(\vec{r})$ is its potential energy in the atom and the momentum operator is $\vec{p} = i\hbar\nabla$.

In turn, the perturbation is the one introduced by the XUV field, which can be expressed as [8]:

$$H'(t) = \frac{e}{2m_e c} (\vec{\mathcal{A}}(t) \cdot \vec{p} + \vec{p} \cdot \vec{\mathcal{A}}(t)) + \frac{e^2}{2m_e c^2} \mathcal{A}^2(t), \quad (1.45)$$

where $\vec{\mathcal{A}}(t)$ is the vector potential of the electromagnetic field.

Since the XUV field is weak because of the low efficiency in the HHG process, $\mathcal{A}^2(t) \approx 0$. After applying the Coulomb gauge, $\nabla \cdot \vec{\mathcal{A}}(t) = 0$, one finally obtains

$$M_{f,i} = \frac{e}{m_e c} \langle \Psi_f | \vec{\mathcal{A}}(t) \cdot \vec{p}(t) | \Psi_i \rangle \quad (1.46)$$

Going back to Eq.(1.42), the phase of sideband oscillations can be decomposed in $\Delta\phi = \Delta\phi_A + \Delta\phi_{XUV}$. $\Delta\phi_A$ is the atomic phase difference between the two-photon transition matrix elements, small and precisely obtained from well established theory [1]. In turn, $\Delta\phi_{XUV}$ stands for the phase difference between two consecutive harmonics, mostly attributed to the XUV group delay [32]: $\Delta\phi_{XUV} = \phi_{q+1} - \phi_{q-1}$. Therefore, by experimentally measuring the intensities of the sidebands as a function of the delay τ , and fitting a cosine to this, $\Delta\phi$ can be inferred. Knowing $\Delta\phi_A$, the phase difference between consecutive pairs of harmonics can be directly calculated. Arbitrarily assigning the phase of one of them to zero for simplicity, and together with the relative harmonic intensities extracted from the PES, the temporal intensity profile of the total field is finally and uniquely determined (see Eq. (1.40)), allowing a reconstruction of the attosecond pulse train.

Chapter 2

Beamline

The upcoming chapter is dedicated to the description of the attosecond beamline used in this thesis, built in a pump-probe configuration which allows for time resolved measurements in the attosecond scale. In general terms, an intense *pump pulse* is employed to trigger a non-equilibrium state on a certain target. A weaker *probe pulse*, phase-related to the latter and sent to the other arm of the interferometer, is later recombined and used to monitor the evolution of the interaction as a function of the optical time delay between the two pulses. For the two-photon ionization measurements taken in this project, both of them came from the output of a Titanium-doped Sapphire (Ti:Sa) laser, which was partly used to generate an attosecond pulse train in the XUV range via high harmonic generation in a gas target (pump pulse), and partly to monitor this process with a much weaker beam (probe pulse). The addition of a home-built NOPA will make it possible to tune the frequency of the probe beam in future experiments. The pump and probe beams are finally recombined by means of a center-hole mirror and focused into a second gas target inside a VMI spectrometer by a toroidal mirror. After ionization, photoelectron kinetic energies and angular distributions are analysed. An XUV spectrometer located at the end of the beamline was also used for the characterization of the resulting harmonics. Bearing in mind the low efficiency of the process and their great absorption by air, all the chambers shown in **Figure 2.1** are kept under special vacuum conditions.

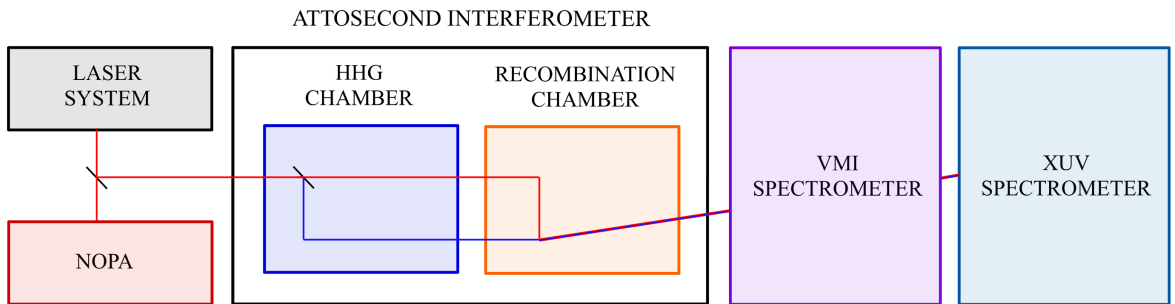


Figure 2.1: General overview of the experimental setup. The output of a Ti:Sa laser system is partially used to feed both the NOPA and the attosecond interferometer. In this work, the probe (red) and pump (blue) beams are splitted at the HHG Chamber and recombined later at the Recombination Chamber. A subsequent focusing into a gas target inside the VMI spectrometer allows for the two-color photoionization experiment, while an XUV spectrometer is used to record the harmonic spectrum.

2.1 The Ti:Sa laser system

Since HHG is a highly nonlinear process, extremely intense sources of light are needed. Given the impossibility of reaching such values with the existing continuous wave (CW) lasers, the alternative comes by the hands of pulsed laser systems. Instead of emitting radiation *continuously*, they concentrate and release great amounts of energy in a shot-to-shot basis.

In this project, the commercially produced FEMTOPOWERTM V CEP4 laser system, from FEMTOLASERS Produktions GmbH, was used. This system delivers ≈ 25 fs pulses with a repetition rate of 1 kHz, at a central wavelength of ≈ 780 nm and a peak power of few hundreds of gigawatts per pulse. It counts with an active CEP stabilization system, and as described in its user manual [33], is divided in four main parts, illustrated in the figure below: CW pump laser, oscillator, amplifier housing (made up by the stretcher, the amplifier and the booster) and compressor.

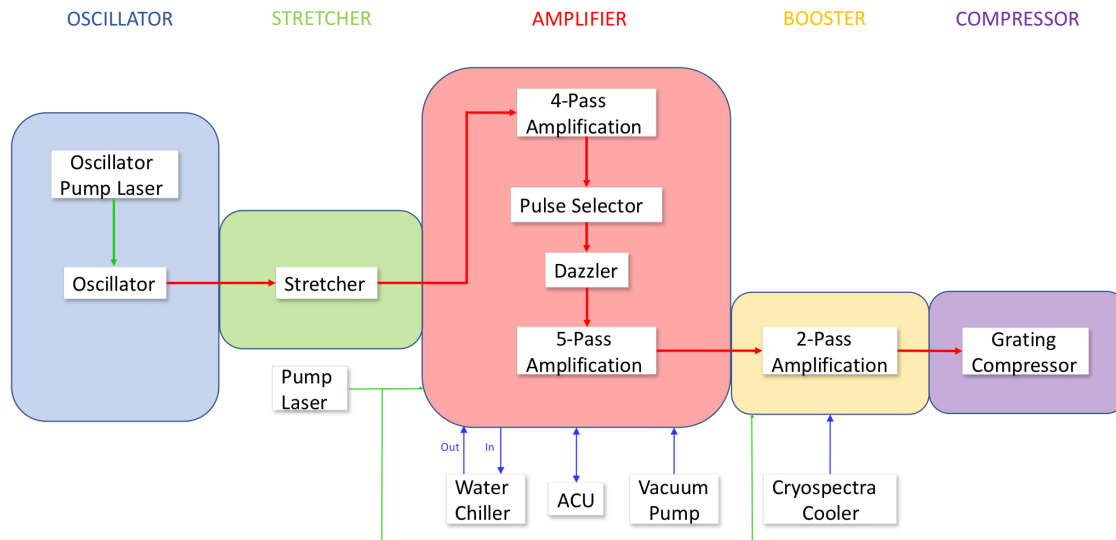


Figure 2.2: Diagram of the laser system. Taken from [34].

In the first place, a mirror-dispersion controlled oscillator generates broadband, ultra-short pulses at approximately 75 MHz repetition rate. This seed beam enters the amplifier housing and gets into a Faraday insulator, in charge of preventing back-reflections and rotating its polarization. Later on, it is sent to a series of Brewster-angled glass blocks that stretch the pulses to a few tens of picoseconds (10^{12} s), reducing their peak power and allowing a safe amplification, always below the damage threshold of the Ti:Sa crystal. This is the so-called *chirped pulse amplification* (CPA) technique, and it is illustrated in **Figure 2.3**. Following this, the beam is sent to the multi-pass pre-amplifier, pumped by a fraction of the kHz-pump laser. Here, it completes a total of nine passes through the first Ti:Sa crystal. After the full MHz-pulse train coming from the seed oscillator completes the first four passes, a single pulse near the maximum of the gain envelope is then selected from the amplified pulse train by a fast-switching Pockels cell, which is shaped both in amplitude and phase by a DAZZLER acousto-optic programmable filter to reduce the effect of gain narrowing¹ in the amplifier and

¹The effect by which the bandwidth of a light pulse gets decreased during amplification, as a

to compensate for third- and fourth- order dispersion. Subsequently, the beam completes the remaining five passes and proceeds to enter the booster, where it gets further amplified to a pulse energy of about 4 mJ after passing twice through another Ti:Sa crystal pumped by the kHz-pump laser. Finally, the amplified pulses are re-compressed to about 60 fs by a grating compressor consisting of two high-efficiency transmission gratings designed to minimize dispersion and a vertical retroreflector.

This laser system is equipped with a couple of active beam-pointing stabilization devices controlled by piezo-actuated mirrors. With the purpose of ensuring a clean environment and to prevent both icing and thermal lensing, the Ti:Sa crystals used in the pre-amplifier and booster stages are kept in vacuum chambers and cooled by water chillers and cryo-refrigerators. Their temperature is measured by an amplifier control unit (ACU) and is of about 235 K for the former, and about 175 K for the latter. The ACU is connected to the main amplifier chamber and keeps track of the pressure in the vacuum chambers as well, in such a way that if an abnormal condition is detected, the kHz-pump laser is automatically switched off.

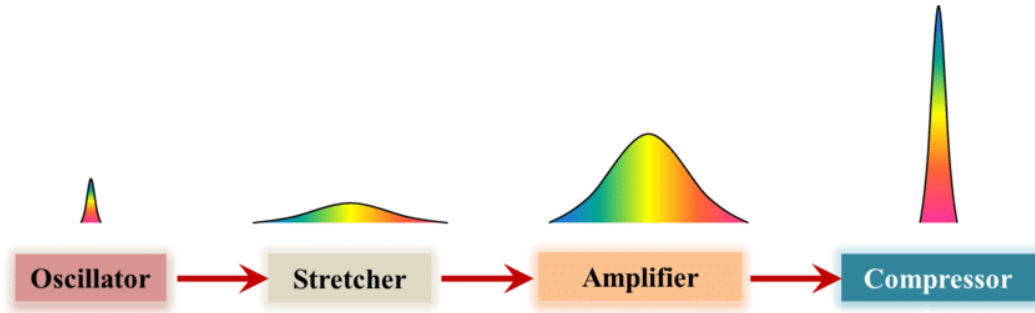


Figure 2.3: Illustration of the CPA technique, for which Gérard Mourou and Donna Strickland were awarded the Nobel Prize in Physics in 2018. Without this method, amplification of ultrashort pulses would be strongly constrained by the low damage threshold of the optics in use. Taken from [36].

2.2 The NOPA

The main author of the noncollinear optical parametric amplifier used in this work is PhD student Muhammad Jahanzeb. It consists of two stages designed to amplify $\lambda_{\text{signal}} = 1200$ nm, with the purpose of later using this signal as the probe pulse in two-color photoionization experiments. In both of the stages, the wavelengths of the pump, signal and idler are therefore:

$$\begin{aligned}\lambda_{\text{pump}} &= \lambda_3 = 780 \text{ nm} \\ \lambda_{\text{signal}} &= \lambda_1 = 1200 \text{ nm}, \quad \text{and} \\ \lambda_{\text{idler}} &= \lambda_2 = \left(\frac{1}{\lambda_3} + \frac{1}{\lambda_1} \right)^{-1} \approx 472 \text{ nm} \quad \text{according to Eq.(1.18),}\end{aligned}$$

consequence of the fact that the central region of the optical spectrum experiences a higher gain than the spectral wings. This is in turn translated into a tendency to increase the pulse duration [35].

where λ_{pump} is the output wavelength of the Ti:Sa laser system. After going through a 50/50 beam splitter (BS1 in **Figure 2.4**), one half of the beam is sent to the attosecond interferometer and the other one to the NOPA setup, as illustrated in **Figure 2.1**. Its spot size is then reduced with a telescope made up by two focusing mirrors. Next, it is lifted to a height of 75 mm by a periscope and reflected into another mirror, which sends it to the next beam splitter, BS2.

First amplification stage

Since the intensity of the seed signal needed to trigger the OPA process must be weak in comparison with the one of the pump, only 10% of the NOPA beam is transmitted through BS2. A $\lambda/2$ waveplate rotates the polarization from parallel to perpendicular to the plane of incidence, with the purpose of later fulfilling the phase matching condition in the nonlinear crystal. After turning its direction of propagation with two 45° mirrors, an attenuator fine tunes its intensity before getting focused into a sapphire crystal plate. WLG takes place, but all wavelengths below 950 nm are blocked right away by another filter. The rest of the (near infrared) spectrum, containing the signal frequency, gets finally projected into the type II, 0.7 mm-thick, BBO crystal by a focusing mirror.

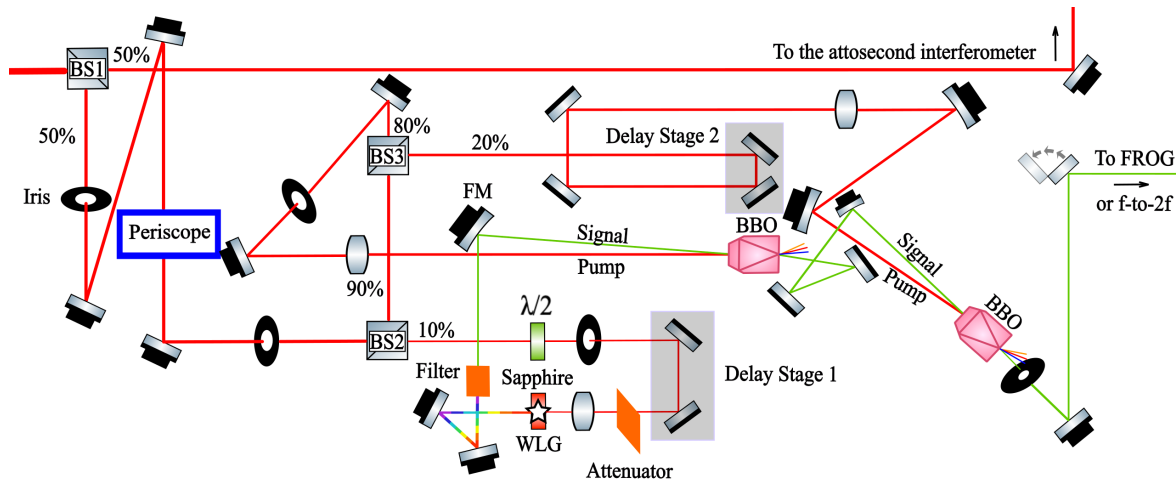


Figure 2.4: Sketch of the home-built NOPA.

The remaining 90% of the NOPA beam gets reflected at an angle of 90°, and pumps both the first and second amplification stages after getting splitted by BS3. The transmitted part of the pump, carrying 80% of its total energy, gets reflected in two mirrors and is finally focused into the BBO crystal by a lens of large focal length, forming an angle α of 3° with the broadband NIR signal described above. At this angle, phase matching is accomplished for $\lambda_{signal} = 1200$ nm.

Optical parametric amplification takes place inside the BBO, and four noncollinear beams make up the output of the process: the idler (represented with orange color in **Figure 2.4**), the weakened pump of the first amplification stage (red), its second harmonic, with $\lambda_{sh} = 400$ nm (blue); and the strengthened signal (represented in green).

Second amplification stage

From the output of the first stage, only the signal is sent to the second BBO crystal of the setup for a second amplification stage.

Regarding the pump, one needs to go back to BS3. The reflected part of the Ti:Sa beam, carrying the remaining 20% of the energy, is guided to another set of mirrors and is finally focused into the second BBO crystal, forming again an angle α of 3° with the signal, set by the phase matching condition.

The final output is also made up by the four noncollinear beams discussed two paragraphs above. In this case, the iris placed right after the crystal for alignment purposes blocks all beams but the signal, which is reflected and exits the NOPA setup. The last component it finds is a flipping mirror, which guides the beam towards the frequency-resolved optical gating (FROG) and f-to-2f setups used for its characterization and described in **subsection 3.3.1**. During the next months after the submission of this thesis, the mirror will eventually be flipped down and the amplified signal will find its way to the attosecond interferometer.

2.3 The Attosecond Interferometer

In order to reconstruct the attosecond pulse train generated via HHG, it was already shown back in Eq.(1.40) that two main parameters are needed: its amplitude and the phase of its constituent harmonics. While the former is directly obtained from the relative intensity of the harmonic comb spectrum, the phase difference between consecutive harmonics is calculated applying the RABBITT technique. As it was explained in the previous chapter, the key point is to make the NIR and the XUV fields interfere between each other. By delaying one with respect to the other, the intensities of the resulting sidebands are made to oscillate in purpose with the goal of extracting $\Delta\phi$ from the interference term $2 \left| \mathcal{M}_{q-1}^{(+)} \right| \left| \mathcal{M}_{q+1}^{(-)} \right| \cos(2\omega\tau - \Delta\phi)$ shown in Eq.(1.42).

The sketch of the attosecond interferometer used in this thesis is illustrated in **Figure 2.5**. It follows a Mach-Zender configuration, and is divided into two chambers under special vacuum conditions: the “HHG Chamber”, where the output of the Ti:Sa laser system is splitted into the two arms of the interferometer to obtain the pump and probe pulses; and the “Recombination Chamber”, where the NIR and XUV fields are recombined and later focused into the interaction point, located in the next component of the setup (the VMI spectrometer).

HHG Chamber

The Ti:Sa output that feeds the interferometer is first divided with an 80/20 beam splitter (BS1).

The reflected beam takes 80% of this energy and gets reflected into a series of plane mirrors. After getting splitted again by a 50/50 beam splitter (BS2) placed for observation purposes, it is later focused by focusing mirror (FM_{pump}) into the gas jet coming from the nozzle, where high harmonics are generated. In this thesis, argon (Ar) was used, as well as a $50 \mu\text{m}$ nozzle. The resulting XUV radiation, from now on known as

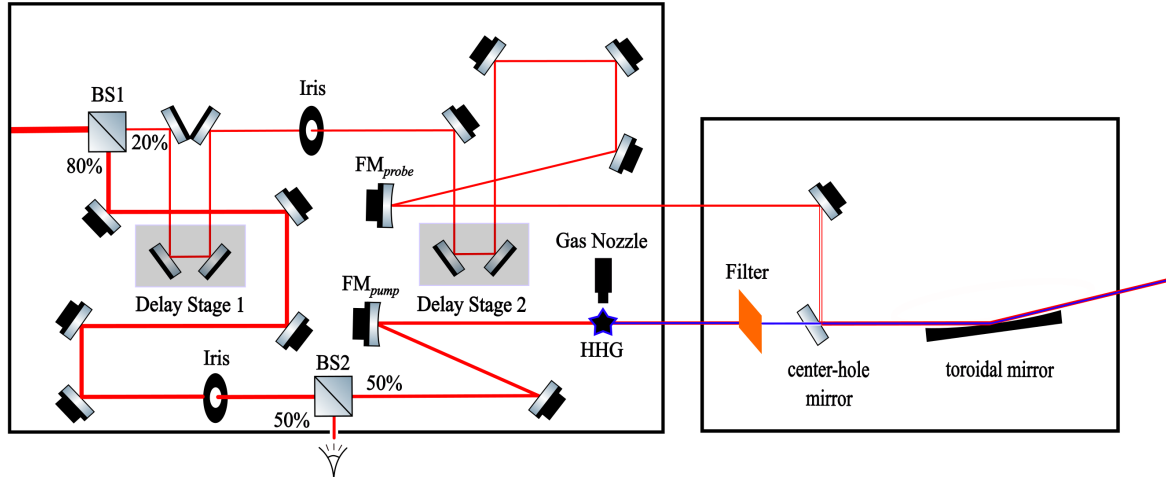


Figure 2.5: The attosecond interferometer, being the one on the left the HHG Chamber, and the one on the right, the Recombination Chamber.

the pump beam and represented with blue color in the diagram, copropagates along with the NIR field towards the Recombination Chamber.

The probe beam takes the remaining 20% of the input and goes through the first delay stage of the setup, made up by two 45-degree mirrors placed in front of a piezo-controlled retroreflector. Then it goes through the second delay stage, and subsequently, gets reflected into a set of three mirrors to finally reach the focusing mirror FM_{probe} , which guides it into the Recombination Chamber, parallel to the pump beam.

Recombination Chamber

The pump beam finds first a motor-controlled rotatory wheel from which different filters can be selected in order to block the original NIR field copropagating with the harmonics. There are two aluminum (Al) and one tin (Sn) filters available, as well as an additional fourth position which opens the possibility of not performing any filtering operation at all. After it, the XUV pulse train reaches a center-hole mirror and passes through. In turn, the probe beam, entering from above in **Figure 2.5**, gets first reflected by a flat mirror and meets the pump pulse at the center-hole mirror. Thereby, the final beam is composed of two well differentiated parts: the XUV pulse train (inner beam), and the NIR field (outer beam). These pulses are phase coherent, which is an essential requirement in order to apply the RABBITT technique. In the last place, the copropagating beams arrive at the toroidal mirror and get focused into the interaction point inside the VMI spectrometer, where another gas target is located and two-color photoionization takes place.

Before moving on to the next component of the setup, three key questions must be addressed:

1. The foci of both the pump and the probe pulses must coincide exactly at the interaction point inside the VMI spectrometer². Nevertheless, one obvious technical obstacle arises if one bears in mind the completely opaque shielding of the

²And given that they are both focused by the toroidal mirror, this is the reason why the probe beam gets also focused before reaching the Recombination Chamber.

device, which besides, is kept in vacuum: verifying their foci is in fact not feasible in a direct way.

2. In order for two light pulses to interfere between each other, they must overlap both in space and time domains. In other words, they must reach the exact same position, at the exact same time. Partial overlapping in both cases allows interference as well, but of course, not in optimal conditions. This can be intuitively understood if one thinks about light pulses as cars: in order for two cars to collide (interfere) between each other, they need to arrive to the same traffic light, at the same time. If they meet slightly before or after, either in space or time, the crash might not be devastating but there will still be some interaction between them.

3. The fringe contrast C is given by

$$C = \frac{I_{max} - I_{min}}{I_{max} + I_{min}} = 2 \frac{\sqrt{I_{pump} I_{probe}}}{I_{pump} + I_{probe}} \quad (2.1)$$

It can be seen that contrast is maximized when $I_{pump} = I_{probe}$, that is, when both beams have the same intensity.

Problem number one was first addressed by extracting the beam out of the beamline through a window located in the arm connecting the Recombination Chamber and the VMI spectrometer. Making use of a beam profiler and measuring the focus position of each beam one at a time, it could be verified that they were not situated at the exact same distance. This was a consequence of the several alignment optimizations of both the laser system and the previous chambers, but could be solved by adjusting their corresponding focusing mirrors. Additionally, they were also made to overlap in space and it was also verified that the distance from the observation window to the foci position was the same as the distance between the former and the interaction point inside the VMI spectrometer.

Next, time overlapping was accomplished by adjusting the optical distance covered by the probe beam, which could be varied by the motorized delay stages shown in **Figure 2.5**. After a coarse scanning, a much more precise one was performed using the piezo-controlled retroreflectors, which provided an accuracy in the micrometer scale. Interference fringes were first observed by coupling the beam into an optical fiber connected to an Ocean Optics spectrometer, and the final visualization of only one fringe oscillating in time is what indicated perfectly collinear propagation of both of the beams into the VMI spectrometer. Using again the beam profiler, intensity oscillations around a certain micrometer range were finally seen.

Problem number three was faced by adjusting the diameter of the iris corresponding to each path.

2.4 The VMI spectrometer

Figure 2.6 illustrates the VMI spectrometer used in this thesis. The two-color beam from the interferometer is focused in the interaction point. Perfectly perpendicular to it and following the red arrow, an argon gas jet descends and photoionization takes place. The resulting photoelectrons are then guided through the TOF tube by the repeller, extractor and ground electrodes (electrostatic lenses 1, 2 and 3 were not used the day of the experiment). The corresponding 3D NS is projected on the 2D MCP surface, and the consequent electron avalanche is accelerated towards a phosphor screen (not shown in the figure) by a potential difference. The CCD camera finally records the fluorescent signal and sends it to the PC. For a more detailed description of its working principle, please refer to **subsection 1.3.1**.

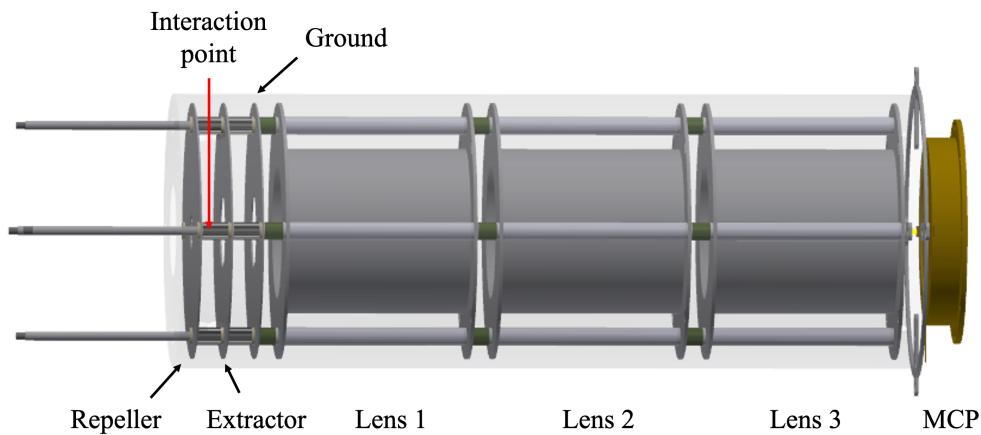


Figure 2.6: Sketch of the VMI spectrometer.

The optimization of the device was performed before the start of this thesis, and led to a voltage of -5 kV for the repeller, -3.34 kV for the extractor, around 2 kV for the MCP and 2.4 kV for the phosphor screen.

A special feature worth to be mentioned is the Behlke switch. It happens that after photoionization, the generated photoelectrons and the ions they leave behind have opposite charges, and correspondingly, get accelerated towards opposite sides under the same potential difference. While the former are guided through the subsequent holey electrodes, the latter hit the repeller with a kinetic energy able to pull electrons apart. If not prevented somehow, they also get accelerated towards the flight tube and projected into the MCP and phosphor screen, contaminating the measurement. What the Behlke switch does is rapidly *switching* the MCP power supply to a low enough value after detection of the main photoelectrons, in such a way that the signal coming from the secondary electrons does not get amplified and the measurement remains reliable.

2.5 The XUV spectrometer

This last element of the beamline was employed to register the spectrum of the generated high harmonics.

Its first component is a reflection grating, in charge of spatially splitting the different harmonics. In this setup, a Hitachi blazed grating, model 001-0640 [37], was used. Designed to maximize the first-order diffraction efficiency for wavelengths in the XUV range (11 nm to 62 nm), it also counts with a concave surface with the purpose of focusing the harmonics and increasing the resolution. Additionally, a thick blocker made of steel and positioned right after it ensures the total block of the zeroth diffraction order.

After a propagation length of 47.5 cm, they are finally recorded by a mobile detection system which allows the recording of different parts of the spectrum. As in the VMI spectrometer, it consists of a MCP, a phosphor screen and a CCD camera directly connected to the PC.

Chapter 3

Measurements, Analysis & Results

The following chapter is dedicated to the presentation, analysis and discussion of the measurements taken in this project.

On the first place, a one-color field experiment was done. After optimizing the high harmonics and analysing their spectrum, the probe beam in the attosecond interferometer was blocked while the XUV radiation was sent inside the VMI spectrometer to ionize an atomic beam of argon. As a result, rings corresponding to different photoelectron kinetic energies were observed. The raw 2D projection of the 3D NS was analysed and Abel-inverted using the pBASEX-Method, and finally the corresponding photoelectron spectrum was extracted.

Then, a two-color field experiment was performed. The probe beam was unblocked and sent with the pump pulse to the VMI spectrometer, as shown in **Figure 2.5**. A telescope was built and installed with the purpose of decreasing the size of the beam. As a result of photoelectron interaction with the NIR field, sidebands appeared halfway between the harmonic peaks. They were visible both in the Abel-inverted images and in the corresponding photoelectron spectrum, which was compared with the one obtained in the one color-field experiment.

To conclude and with the presence of Dr. Cristian Manzoni, Senior Researcher at IFN-CNR, the setup of the NOPA was optimized and its output characterized. The maximum achievable power of the signal using two BBO crystals with different thicknesses was compared. From their corresponding spectra, the Fourier transform limit was first calculated, and then, the actual pulse duration of the optimal configuration was measured using a frequency-resolved optical gating (FROG) setup. The intensity stability of the amplified signal was analysed using a photodiode and an oscilloscope. With the purpose of measuring its CEP stability, an f-to-2f interferometer was built, and the Fourier Transform Spectral Interferometry (FTSI) algorithm was implemented.

3.1 One color-field experiment: XUV only

Before the realization of the two-color experiment and with the purpose of testing the optimization of the setup alignment and VMI spectrometer, a one color-field experiment was first performed.

3.1.1 Optimization and characterization of the harmonics

The probe beam in the HHG Chamber (see **Figure 2.5**) was blocked, and the high harmonics generated by the other arm on the argon target were optimized on the first place. In order to do so, the XUV spectrometer was employed. After blocking the residual NIR field with an aluminum filter placed inside the Recombination Chamber, the voltage of the MCP was set to 0.7 kV, while the one of the phosphor screen to 2.5 kV. With the CCD camera, harmonics were observed in real time. By slightly adjusting the position of the gas nozzle and the DAZZLER acousto-optic programmable filter to compensate for higher order dispersion effects, the amplitude and amplification gain were eventually maximized. The power of the beam was also varied and finally set to 777 mW before the interferometer. In order to determine the order of the first visible harmonic, one needs to compare the spectrum to the transmission curve of the aluminum filter used to block the NIR parent field, shown in **Figure 3.1**. Additionally and due to the symmetry inversion properties of noble gases, only odd harmonics separated by twice the energy of the NIR parent field ($E_{NIR} = \frac{hc}{\lambda_{NIR}} \approx 1.59$ eV) are visualized, as anticipated in **subsection 1.4.1**.

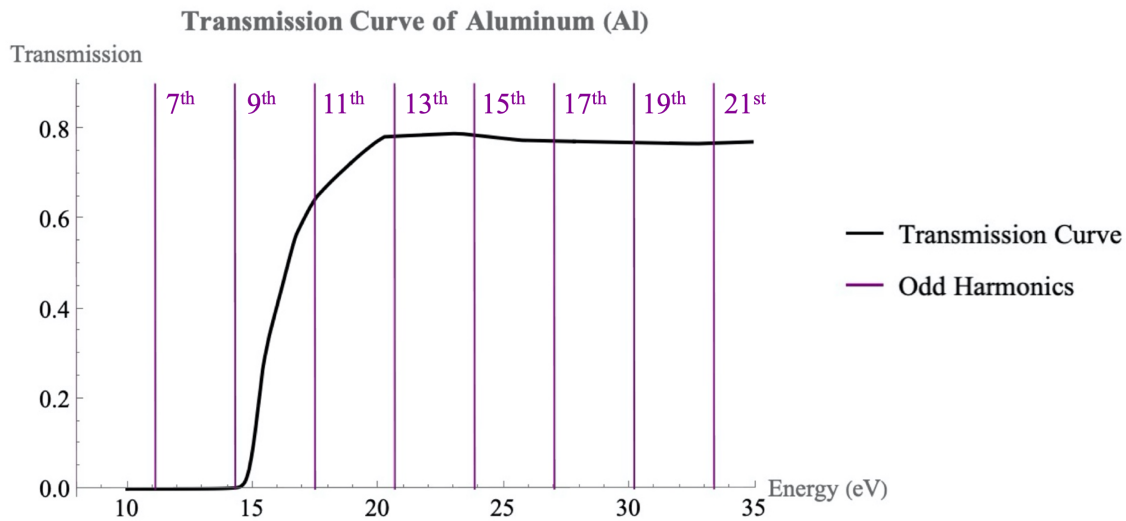


Figure 3.1: Transmission curve of the 99 nm-thick aluminum filter used to block the NIR field as a function of the energy (black). Data has been extracted from [38]. Odd harmonics are represented by purple, vertical lines.

In view of this graph, one could be tempted to assert that the first (partially) transmitted harmonic is the eleventh one. Although this is indeed correct, it is not the first visible harmonic in the XUV spectrometer because it is out of the range for which the blaze grating is optimized (11 nm $\sim$ 62 nm, [37]):

$$\lambda_{11th} = \frac{hc}{11 \cdot E_{NIR}} = \frac{780 \text{ nm}}{11} \approx 71 \text{ nm} \notin [11 \text{ nm}, 62 \text{ nm}], \text{ while}$$

$$\lambda_{13^{th}} = \frac{hc}{13 \cdot E_{NIR}} = \frac{780 \text{ nm}}{13} \approx 60 \text{ nm} \in [11 \text{ nm}, 62 \text{ nm}]$$

The harmonic spectrum obtained the day of the one and two color-field experiments is plotted in **Figure 3.2** (right). Up to nine harmonics are visible:

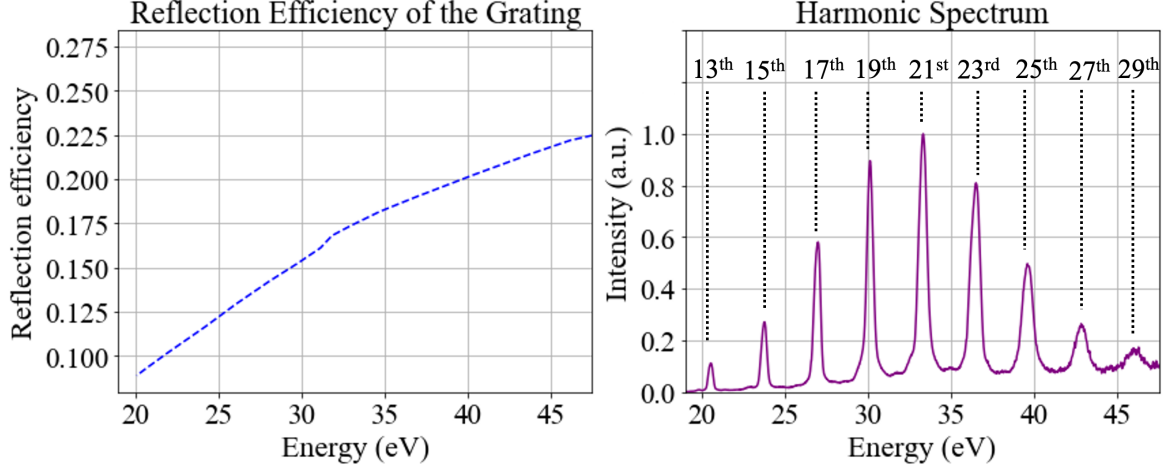


Figure 3.2: Left: reflection efficiency of the grating in the XUV spectrometer, taken from the specifications of the product. Right: Harmonic spectrum obtained the day of the experiment. Orders from the 13th to the 29th are visible.

On the one hand, the reason why their intensity seems to increase and reach a peak at harmonic 21st is explained by the reflection efficiency of the grating, plotted in **Figure 3.2** (left). It has a minimum value for the lowest energies. On the other hand, the typical structure of a high harmonic spectrum is divided in three parts, as depicted in **Figure 3.3**: the perturbative region, where intensity decreases with increasing harmonic; the plateau, where the intensity does not depend significantly on the energy of the harmonic; and the cutoff, where intensity finally decreases with increasing order of harmonic. This energy cutoff can be calculated according to the formula

$$E_{cutoff} = I_p + 3.17U_p, \quad \text{where} \quad U_p = \frac{e^2 I}{2c\epsilon_0 m \omega^2} \quad (3.1)$$

is the mean kinetic energy of an electron in the external electric field, averaged over an optical cycle. In this term, e is the electrical charge of the electron, I is the intensity of the incident field, c is the speed of light, ϵ_0 is the permittivity in vacuum, m is the mass of the electron and $\omega = 2\pi c/(780 \text{ nm})$ is the angular frequency of the incident field. Since 777 mW were measured before the attosecond interferometer, and according to **Figure 2.5** 80% of the energy is reflected by BS1 and 50% of that by BS2, the incident average power should in this case be of about 310.8 mW. Dividing this value by the repetition rate of the Ti:Sa laser system ($f_{rep} = 1 \text{ kHz}$), one obtains the average pulse power. Dividing again by the pulse duration of the Ti:Sa output ($\tau_{Ti:Sa} \approx 30 \text{ fs}$), the peak power per pulse is obtained. To determine the intensity, we divide by the area of the beam at the focus, which can be approximated by $\pi \cdot (55 \mu\text{m})^2$. Plugging this values into Eq.(3.1) yields $E_{cutoff} \approx 35 \text{ eV}$, and this is why intensity starts decreasing approximately after harmonic 21st. To summarize,

$$I = \frac{\text{Peak Power}}{\text{Area}} \approx \frac{\text{Average Power}/(f_{rep} \cdot \tau_{Ti:Sa})}{\pi r^2} = \frac{310.8 \text{ mW}}{1 \text{ kHz} \cdot 30 \text{ fs} \cdot \pi \cdot (55 \mu\text{m})^2} \Rightarrow$$

$$\Rightarrow I \approx 1.09 \times 10^{18} \text{ W/m}^2 \quad \Rightarrow \quad U_p \approx 6.2 \text{ eV} \quad \Rightarrow \quad E_{\text{cutoff}} \approx 35 \text{ eV}$$

Note however that this is just a rough calculation. In order to precisely compute the exact result, one would also need to consider the energy losses on the various mirrors of the beam path, the approximately Gaussian nature of the pulses, etc.

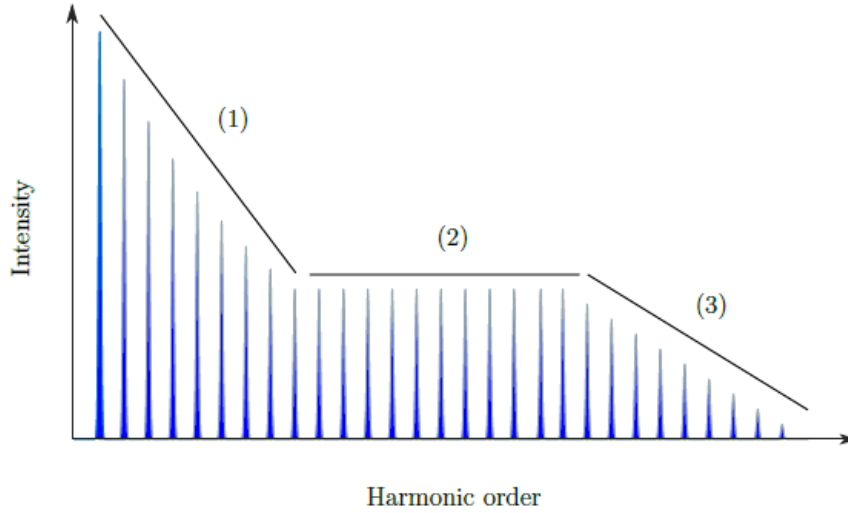


Figure 3.3: Typical spectrum resulting from the HHG process. Region (1) corresponds to the perturbative one, region (2) to the plateau and region (3) to the cutoff. Taken from [39].

3.1.2 VMI measurements

Next, attention was shifted towards the VMI spectrometer. An atomic beam of argon was sent to the interaction point, and the following voltages were applied to the different components of the spectrometer:

$$\begin{aligned} V_{\text{repeller}} &= -5 \text{ kV} \\ V_{\text{extractor}} &= -3.335 \text{ kV} \\ V_{\text{MCP}} &= 1.8 \text{ kV} \\ V_{\text{phosphor}} &= 5 \text{ kV} \\ V_{\text{lens 1}} &= V_{\text{lens 2}} = V_{\text{lens 3}} = 0 \text{ V} \end{aligned}$$

As a consequence of the ionization of argon by the incoming XUV radiation, a symmetric, circular distribution of photoelectrons projected onto the MCP was observed, as illustrated in **Figure 3.4**. This image shows the result of a one-hour long scan. A background measurement, where the atomic beam was cut off, was taken in a different scan and subtracted from the latter. From the figure, two important observations can be made:

1. The most evident one is that there appears to be six different rings, each of them associated to a different photoelectron kinetic energy. Particularly, inner rings correspond to lower harmonics: after overcoming the ionization potential of argon, $I_p^{\text{Ar}} = 15.76 \text{ eV}$ [40], and given that harmonic n has n times the energy of the parent

NIR photon, they are left with less kinetic energy to transfer to the photoelectrons compared to higher-order harmonics, and as a result, photoelectrons get projected in rings of lower radii. **Table 3.1** summarizes this information. Although the eleventh harmonic cannot be detected with the XUV spectrometer, it is indeed the first one able to ionize the argon atoms:

$$E_{11^{th}} = 11 \cdot E_{NIR} = 11 \cdot \frac{hc}{780 \times 10^{-9}\text{m}} = 17.485 \text{ eV} > 15.76 \text{ eV} = I_p$$

Therefore, the most inner ring corresponds to ionization by the eleventh harmonic.

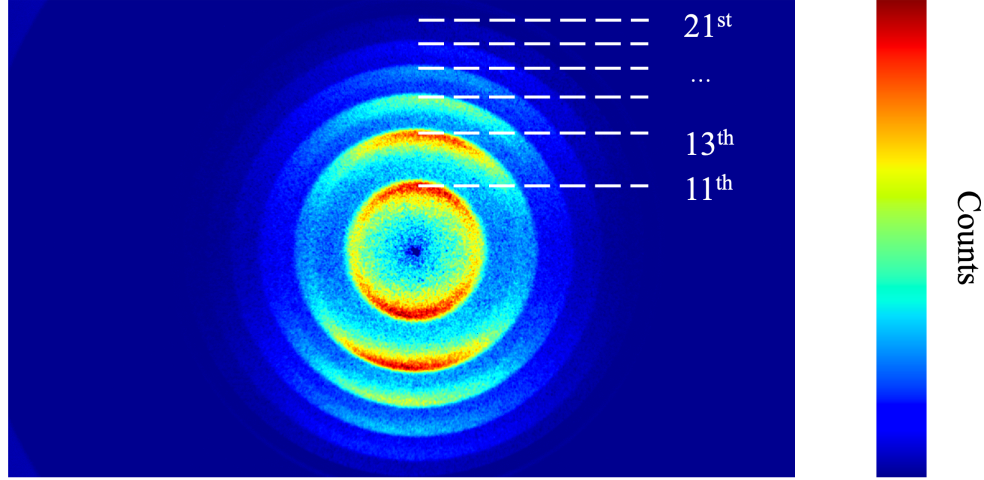


Figure 3.4: 2D projection of the 3D NS in the one color-field experiment. Redder areas show higher concentration of photoelectrons. Each of the dashed lines highlights the ring corresponding to ionization by the odd harmonic indicated next to them.

Table 3.1: Energies of the detected odd harmonics (left) and kinetic energies of the resulting photoelectrons (right).

Energy of n^{th} -Harmonic (eV)	Photoelectron Kinetic Energy (eV)
$11 \cdot E_{NIR} = 17.48$	$17.48 - I_p^{Ar} = 1.72$
$13 \cdot E_{NIR} = 20.66$	$20.66 - I_p^{Ar} = 4.90$
$15 \cdot E_{NIR} = 23.84$	$23.84 - I_p^{Ar} = 8.08$
$17 \cdot E_{NIR} = 27.02$	$27.02 - I_p^{Ar} = 11.26$
$19 \cdot E_{NIR} = 30.20$	$30.20 - I_p^{Ar} = 14.44$
$21 \cdot E_{NIR} = 33.38$	$33.38 - I_p^{Ar} = 17.62$

2. A higher density of photoelectron population is found both in the upper and lower parts of the rings than on the sides. As introduced back in **subsection 1.3.2**, this is a consequence of irradiating the atomic beam with a linearly polarized laser beam (note however the slight tilt to the right, which indicates that the polarization is not perfectly perpendicular to the VMI axis).

The Abel-inverted image was obtained with the “pBasexl” software by Luka Pravica *et. al*, which applies the pBASEX algorithm,

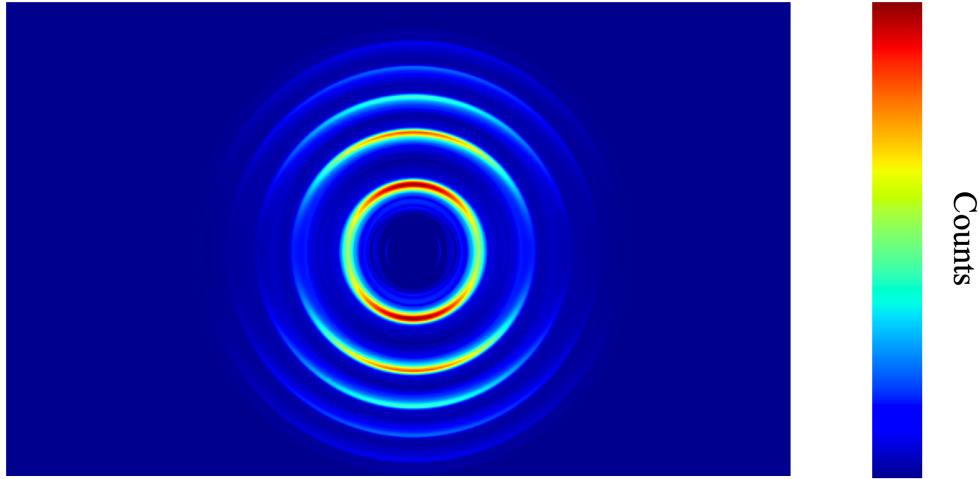


Figure 3.5: Abel-inverted image of the one color-field experiment.

Finally, the corresponding photoelectron spectrum was extracted and presented in **Figure 3.6**. In agreement with the interpretation given above, six peaks at approximately the photoelectron kinetic energies estimated in **Table 3.1** are visible. In order to explain why their intensity decreases with photoelectron kinetic energy, it is useful to take into account how the total absorption cross section of argon atoms varies with ionization energy. As seen in the table below:

Table 3.2: Total absorption cross sections of different photoionization energies provided by [41, 42].

Ionization Energy (eV)	Absorption Cross Section (Mbarn)
15.0	16.7
20.0	50.1
21.2	38.0
25.0	13.5
26.9	8.1
30.0	3.6
35.0	1.5

it reaches a peak at 20 eV and decays rapidly as the ionization energy varies from this value. In other words, argon atoms absorb less efficiently radiation lower or greater than 20 eV (corresponding to harmonic $\approx 13^{\text{th}}$). With the purpose of obtaining a more precise estimation of the absorption cross sections of harmonics 11^{th} and 13^{th} , a linear interpolation was made. Results are shown in **Table 3.3**. Comparing now with **Figure 3.10**, although the absorption peak of harmonic 11^{th} is indeed slightly higher, the one of harmonic 13^{th} is much broader. The area under the curve (total absorption) is thus still greater than in the former case, in agreement with this approximation, and this tells us that photoelectrons are emitted within a certain range in kinetic energy, rather than a well-defined value.

Table 3.3: Estimated absorption cross sections for harmonics 11th and 13th, calculated using a linear interpolation.

	Energy (eV)	Absorption Cross Section (Mbarn)
11 th Harmonic	17.48	33.3
13 th Harmonic	20.66	43.4

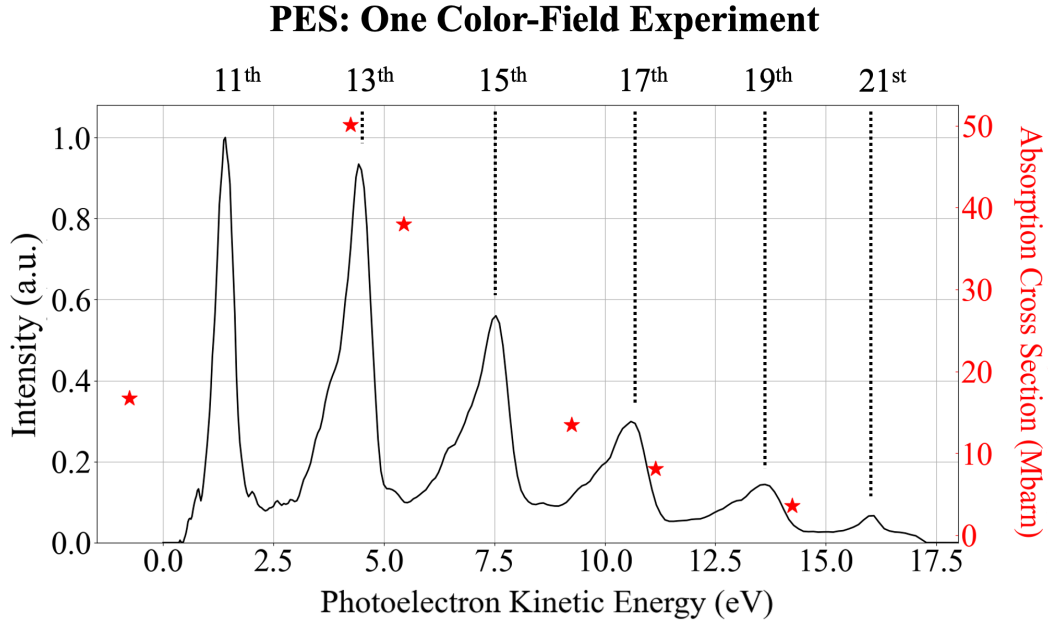


Figure 3.6: Normalized PES of the one color-field experiment (black). Each of the intensity peaks corresponds to ionization by the harmonic indicated above. The red stars are the absorption cross sections of argon atoms as a function of photoelectron kinetic energy (i.e. total energy minus I_p^{Ar} , see Eq.(1.41)).

Some relevant contributions to the total broadening of the lines are:

1. the natural bandwidth of the beam used to create the harmonics and its pulse duration,
2. the Doppler broadening, since different velocities of the emitted photoelectrons result in different Doppler shifts,
3. the fact that the XUV radiation does not ionize at a single point, but rather before and after the focus as well,
4. and space charge effects, since negatively charged photoelectrons repel each other from shot to shot.

Pressure broadening, related to collisions between atoms both in the gas target in the HHG process and in the atomic beam of argon inside the VMI spectrometer, are not that important because gas nozzles are designed to minimize this effect.

3.2 Two color-field experiment: XUV + NIR

The next step in our pursuit for applying the RABBITT technique was the realization of the two color-field experiment, where the probe beam was unblocked and recombined with the attosecond pulse train. With the goal of later establishing reliable comparisons, measurements were taken right after performing the one color-field experiment, so laboratory conditions (power, harmonic's spectrum and VMI settings) remained similar. Nevertheless, an important technical issue had to be faced before the eventual observation of sidebands.

3.2.1 Construction of an auxiliar telescope

Measurements of the probe beam power at different stages inside the attosecond interferometer were taken; a certain loss of energy was anticipated due to the presence of the iris on the probe beam path and the center-hole mirror. **Table 3.4** summarizes the obtained values (see **Figure 2.5**):

Table 3.4: Initial measurements of the probe beam power at different stages of the attosecond interferometer, where

$$\text{Energy loss} = \left(1 - \frac{P_{out} = 31 \text{ mW}}{P_{in} = 168 \text{ mW}} \right) \times 100\%$$

Input power: 1000 mW	
Position	Power (± 3 mW)
After BS1	181
Before Iris	168
After Iris	45
Before center-hole mirror	36
After center-hole mirror	31
Energy loss: 82%	

As seen, a power of 1 W was set at the entrance of the attosecond interferometer the day these measurements were acquired. The first drop is accounted for the beam splitter, which in principle should transmit 20% of the incoming energy. Although the measured value is slightly lower, this is not the main issue¹: a loss of energy of 82% was detected from this point to the output of the center-hole mirror. This is explained by the fact that since the NIR beam of the other arm of the interferometer has to be focused to create harmonics, the probe beam also needs to be focused so that they cover the same optical path length before reaching the toroidal mirror, where they both get refocused on the interaction point inside the VMI spectrometer. Given that the focal

¹This could be explained by losses of energy on the optics located between the position where 1000 mW were measured and the beam splitter, e.g. the periscope used to guide the beam into the interferometer. It could also be due to a defect on the beam splitter, or alternatively, on the power meter. In the latter case, the same systematic error would affect measurements in the same way, so the reliability of this analysis would not get influenced by it.

length of FM_{probe} is 25 cm and the distance from this mirror to the flat one inside the Recombination Chamber is larger, the probe beam reaches the latter with a larger spot size. In this case, it was such that the total beam size was greater than the surface of the mirror by the time it hit it. In order to avoid this, the corresponding iris inside the Recombination Chamber had to be closed so much that energy decreased by a factor of approximately 3/4. The solution was therefore to reduce the size of the beam by means of an auxiliary telescope placed before the entrance to the attosecond interferometer. Instead of using two convergent lenses separated by a distance of $f_1 + f_2$ (where f_1 and f_2 are the focal lengths of lenses 1 and 2, respectively), one convergent lens (L_1) and another divergent one (L_2) were employed with the purpose of avoiding focusing the laser in the region in between these optics and trigger third-order nonlinear effects such as the ones described in **section 1.2** that would alter the spectral properties. To accomplish a collimated output and since the size of the beam was desired to be reduced to about 1/2 of its original size, the divergent lens was placed so that its image focal plane, F'_2 , coincided with the position of L_1 , as illustrated in **Figure 3.7**.

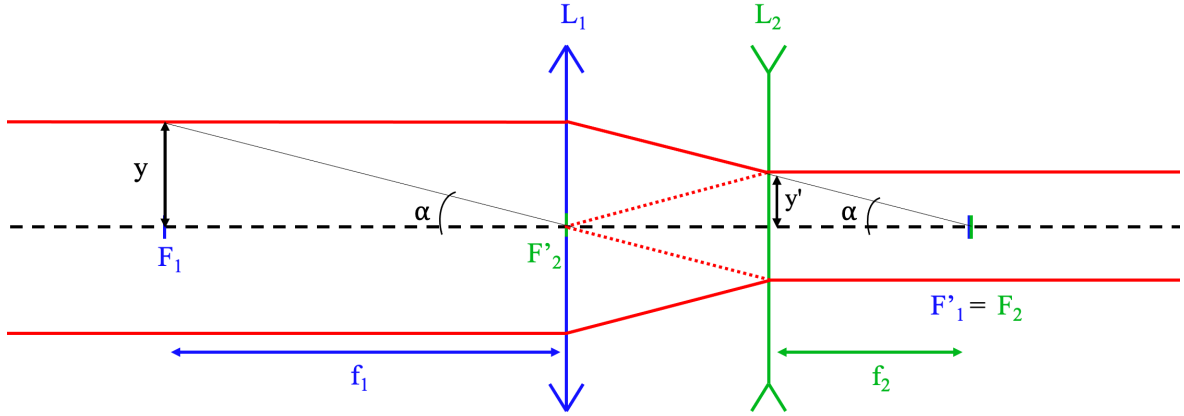


Figure 3.7: Sketch of the telescope used to reduce the size of the beam (red color): L_1 (blue color) represents the convergent lens, while L_2 (green color) represents the divergent lens. F_i , where $i = 1, 2$, are the front focal planes, while F'_i are the corresponding back focal planes; f_i represent the focal lengths and y and y' are the input and output beam radii, respectively. The thin lens approximation is being applied.

From the triangle similarity, it can be seen how the ratio between focal lengths determines the ratio between the input and output beam radii:

$$\frac{y}{f_1} = \frac{y'}{f_2} \quad \Rightarrow \quad \frac{f_1}{f_2} = \frac{y}{y'} \stackrel{!}{=} 2$$

Consequently, a divergent lens of focal length $f_2 = (1/2)f_1$ was finally installed.

Once the telescope was ready to be used and the beam size was effectively reduced, measurements shown in **Table 3.4** were repeated in this new condition. Results are shown in **Table 3.5**:

Table 3.5: Measurements of the probe beam power at different stages of the attosecond interferometer after the installation of the telescope. The energy loss has been calculated the same way as in **Table 3.4**.

Input power: 865 mW	
Position	Power (± 3 mW)
Before Iris	128
After Iris	76
Before center-hole mirror	70
After center-hole mirror	60
Energy loss: 53%	

Note how the input power measured before the attosecond interferometer was lower in this case due to the change of temperature throughout the day and consequent slight misalignment of the laser source. Nevertheless, it is striking how even in this condition the power measured after the center-hole mirror was twice the one obtained before, decreasing the relative loss of energy significantly.

3.2.2 VMI measurements

With this improvement and under the experimental conditions described in the one color-field experiment, another one hour-long acquisition analogous to the one shown in **Figure 3.4** was taken. Sidebands appearing between harmonics were finally observed. Also, a hot spot was visible in the origin, which could be an ionization process resulting from the now too-strong NIR field:

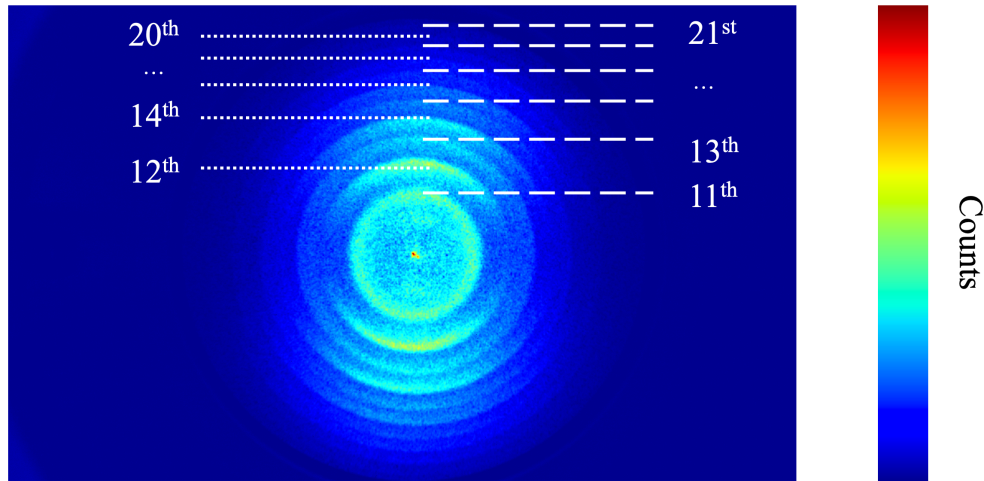


Figure 3.8: 2D projection of the 3D NS in the two color-field experiment. Redder areas show higher concentration of photoelectrons. Each of the dashed lines highlights the ring corresponding to ionization by the odd harmonic indicated on the right, while each of the dotted lines highlights the ring corresponding to the sideband indicated on the left.

Making use of the “pBasex” software, the Abel-inverted image was obtained:

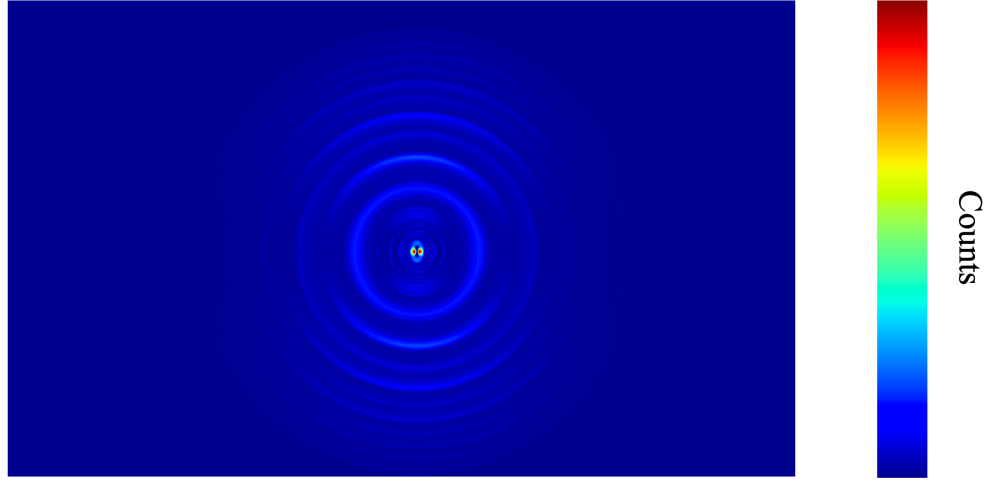


Figure 3.9: Abel-inverted image of the two color-field experiment. A total of eleven different rings, corresponding to the five different harmonics visualized in **Figure 3.9** and their associated sidebands, are visible.

To conclude, the corresponding PES is presented in the figure below:

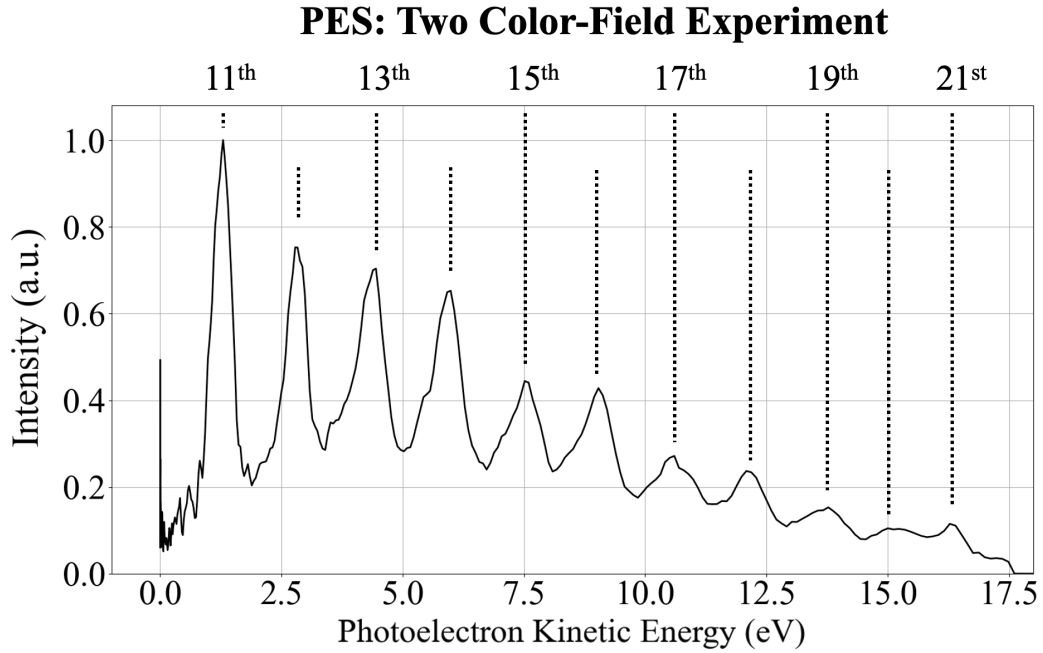


Figure 3.10: Normalized PES of the two color-field experiment (black). Sidebands between each intensity peak corresponding to ionization by the harmonic indicated above appear as a result of photoelectron interaction with the parent NIR field.

Intensity peaks in between the original ones caused by the harmonics are visible. They correspond to sidebands, and they are the result of interaction with the parent NIR field with fundamental, angular frequency ω . In its presence, photoelectrons can absorb or emit a certain number NIR photons, acquiring kinetic energies which are even multiples of $\hbar\omega$. It is worth to note how sideband q is even more intense than harmonic $q + 1$,

which suggests that the input power (777 mW) was so high that multiphoton transitions were taking place, i.e. not two but various quantum paths were interfering between each other. Consequently, the second-order perturbation theory would not be suitable in this situation, and Eq.(1.42) would no longer hold for the accurate description of the intensity oscillations of sidebands as a function of the delay between the XUV and NIR fields. Thereby, taking into account that in practice it is easier to set a lower input power rather than implementing more complex calculations, the next step of the experiment should be finding the minimum NIR intensity for which sidebands are visible. The closer to this value, the more precise will be the reconstruction of the attosecond pulse train originated in the HHG process.

3.3 Optimization of the NOPA

3.3.1 Power, spectrum & Fourier limit

The first task in the optimization of the NOPA was the realignment of the setup, following a step-by-step procedure. With the purpose of obtaining the maximum achievable energy for the amplified signal without exceeding the damage threshold of the BBO crystals, the Ti:Sa total output power was kept at ≈ 1.47 W, 50% of which fed the NOPA.

Regarding the first stage, it was verified how the spectrum of the white light depends strongly on the intensity focused into the sapphire crystal (regulated in the setup by the intensity attenuator, see **Figure 2.4**).

In optimal conditions and with $(34 \pm 3)\mu\text{J}$ of pump energy, a value of $(1 \pm 0.5)\mu\text{J}$ was measured for the amplified signal. Uncertainties have been estimated according to the measurement fluctuations observed in the laboratory. **Figure 3.11** (left) shows the obtained spectrum. It peaks at $\lambda \approx 1200$ nm, although its center of mass (COM) is slightly shifted towards a greater wavelength because of the slight asymmetry observed in the spectrum. This is due to self-steepening effects in the WLW process, as anticipated in **Practical generation of white light**. The COM has been calculated according to

$$\text{COM} = \frac{\sum_i \lambda_i \cdot I_i}{\sum_i I_i} \quad (3.2)$$

In addition, a width $\Delta\lambda$ at full width at half maximum (FWHM) of 169 nm has been estimated.

As a consequence of the Fourier transform, the duration and the spectral width of a pulse are related to each other by the so-called time-bandwidth product:

$$\Delta f \cdot \tau \geq \mathcal{C}, \quad (3.3)$$

where Δf is the frequency bandwidth, τ is the pulse duration and $\mathcal{C}$ is a constant which depends on the shape of the pulse. Assuming a Gaussian-shaped pulse, $\mathcal{C} \approx 0.441$. The time-bandwidth product is at its minimum for the lower limit for the pulse duration which is possible for a given spectrum, that is, when equality in the above expression is reached. If that is the case, the light pulse is said to be *Fourier-limited*: it has a frequency-independent spectral phase, and thus corresponds to a pulse with no chirp. A first estimation of this lower limit was calculated using Eq.(3.3) and the bandwidth $\Delta\lambda$ determined at FWHM:

$$\begin{cases} f_{\max} = \frac{c}{(1200 \text{ nm} - \frac{\Delta\lambda}{2})} \\ f_{\min} = \frac{c}{(1200 \text{ nm} + \frac{\Delta\lambda}{2})} \end{cases} \Rightarrow \tau = \frac{\mathcal{C}}{\Delta f} = 12.5 \text{ fs}, \quad (3.4)$$

where c is the speed of light.

A more realistic result was accomplished by the implementation of a numerical algorithm. Keeping in mind the quadratic relationship between intensity and electric field,

the square roots of the measured intensities were first taken. Then, the (Inverse) Discrete Fourier Transformation (IDFT) was applied in order to express the electric field in the time domain:

$$E(t) = \sum_i E_i(f) \cdot e^{j2\pi f_i t} \Delta f_i, \quad (3.5)$$

where $E_i(f_i)$ is the electric field at frequency f_i , t is the time interval and Δf_i is the difference between frequency f_i and f_{i-1} . This last term takes into account the fact that the number of samples is in this case equidistant in wavelength, but not in frequency.

Finally, intensities as a function of time were calculated by modulus-squaring the resulting electric field as a function of time. The obtained result is presented in **Figure 3.11** (right). The Fourier Limit (FL) was estimated at FWHM.

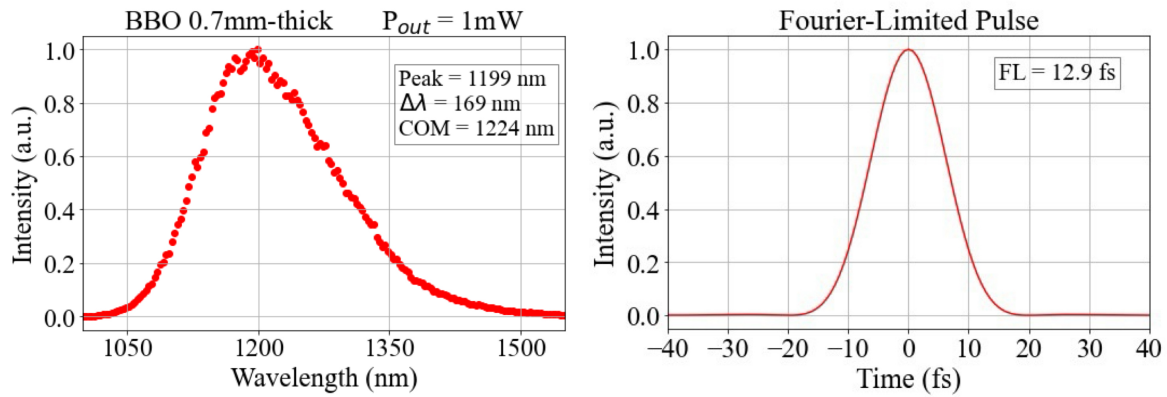


Figure 3.11: Spectrum (left) and Fourier-Limited pulse (right) after the first amplification stage P_{out} stands for “output power”.

Next, attention was shifted to the second stage. The focusing mirror of the signal, as well as the focusing lens and mirror of the pump played a major role in the spatial overlap of both beams. For the temporal part, the effect of delay stage 2 on the phase matching was confirmed by observing how the spectrum shifted to lower or greater wavelengths, and how the output energy dropped. A final energy for the amplified signal of $(60 \pm 3) \mu\text{J}$ was obtained in this case, with $(450 \pm 5) \mu\text{J}$ of pump energy. Results are presented in **Figure 3.12**, where all parameters have been calculated as described in previous paragraphs. In contrast to the first stage, it can be noted how the center-of-mass wavelength is displaced to a lower value now, closer to the ideal $\lambda = 1200 \text{ nm}$. Because phase-matching is accomplished for this wavelength, the spectrum became significantly narrower and consequently the Fourier Limit increased by 5.7 fs.

The performance of a thicker BBO crystal was then analysed and compared to the previous one. Hence, the 1 mm-crystal of the second stage was replaced by a 1.4 mm one, and measurements were repeated in an analogous way. Back in Eq.(1.13), it was already anticipated how gain grows exponentially with interaction length, although dispersion losses become more relevant, too. After optimization, the obtained total energy for the amplified signal was indeed $7.5 \mu\text{J}$ greater; however, the asymmetry in the spectrum was accentuated, and the center of mass wavelength shifted again further than the ideal $\lambda = 1200 \text{ nm}$. Although a shorter bandwidth $\Delta\lambda$ was measured at

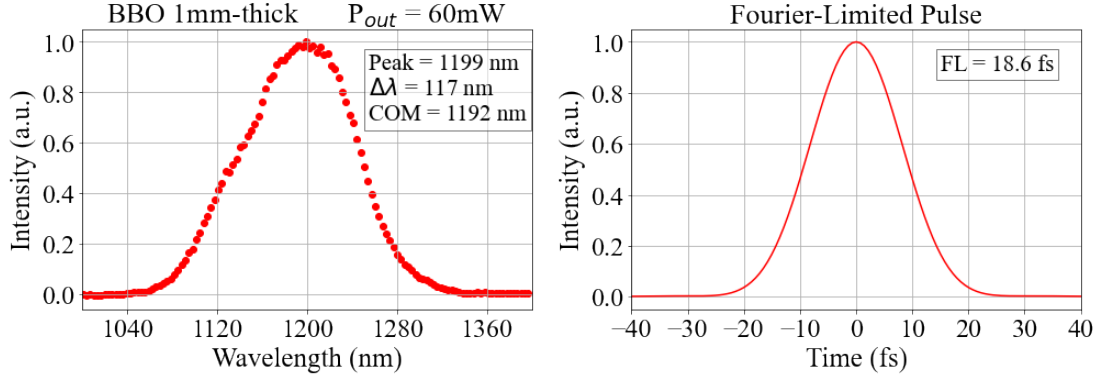


Figure 3.12: Spectrum (left) and Fourier-Limited pulse (right) after second stage amplification.

FWHM comparing with the 1 mm-BBO, the calculated Fourier-Limited-pulse was still slightly shorter because the spectrum was broader at low intensities.

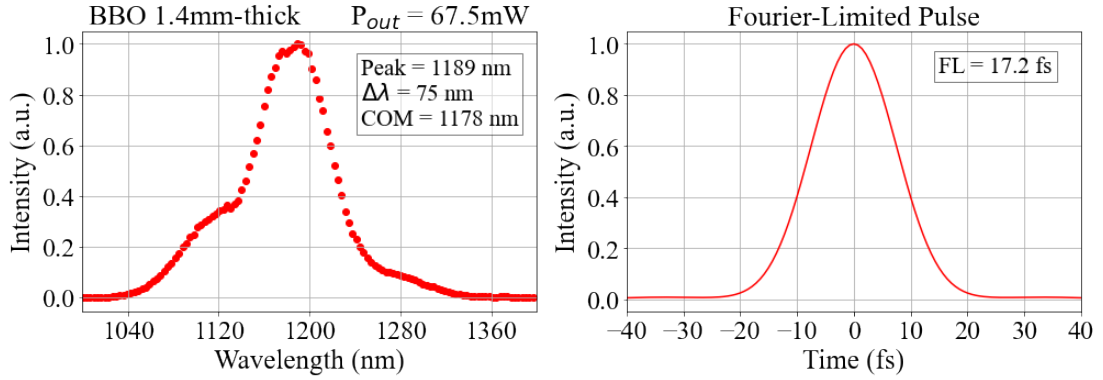


Figure 3.13: Spectrum (left) and Fourier-Limited pulse (right) after second stage amplification, using a thicker BBO crystal.

In order to decide which crystal is more suitable, it is convenient to remember that it will be used as the probe beam in future two color-field experiments:

1.- On the one hand, the longer the probe pulses, the better the resolution of the sidebands will be. This is easy to see by looking at Eq.(1.39). As it will be shown in the next section, already with the 1 mm- thick BBO crystal the NOPA pulses have twice the pulse duration than the one of the Ti:Sa pulses, which means that in this condition the sidebands in **Figure 3.10** will be significantly thinner; resolution will improve (it is true that two sidebands instead of only one will appear between each harmonic, but even with ≈ 30 fs pulses like the ones coming from the Ti:Sa laser system, two sidebands could fit in between each harmonic peak).

2.- On the other hand, the longer the pulse, the less the peak power per pulse². Although the use of the 1.4 mm- thick BBO crystal adds 7.5 mW of probe power because gain grows exponentially with interaction length, it will also increase the pulse duration due to dispersion (the corresponding Fourier limit seems to go against this statement,

²The peak power per pulse of the probe beam is not that critical as in case of the pump beam; however, it still has to be above a certain threshold, as detailed in the next pages.

but this is only because of the asymmetry of the spectrum commented above. It is already seen in **Figure 3.13** how $\Delta\lambda$ decreases significantly at FWHM, and according to Eq.(3.3), this implies a longer pulse duration), and this broadening in the time domain does not compensate the 7.5 mW of gained power. To prove it, and since the actual pulse duration was measured with only one of the crystals, let's calculate the peak power per pulse assuming perfectly compressed and gaussian pulses (i.e. apply the equality in Eq.(3.3), where $\mathcal{C} = 0.441$), and use the bandwidths shown in **Figures 3.12** and **3.13**:

- **1 mm-thick BBO crystal** $\Delta\lambda = 117\text{nm}$ $P_{out} = 60 \text{ mW}$

$$\begin{cases} f_{max} = \frac{c}{(1200 \text{ nm} - \frac{117 \text{ nm}}{2})} \\ f_{min} = \frac{c}{(1200 \text{ nm} + \frac{117 \text{ nm}}{2})} \end{cases} \Rightarrow \tau = \frac{\mathcal{C}}{\Delta f} = 18.1 \text{ fs}$$

$$\text{Peak Power} = \frac{P_{out}}{f_{rep} \cdot \tau} = \frac{60 \text{ mW}}{(1 \text{ kHz})(18.1 \text{ fs})} = 3.3 \text{ GW}$$

- **1.4 mm-thick BBO crystal** $\Delta\lambda = 75\text{nm}$ $P_{out} = 67.5 \text{ mW}$

$$\begin{cases} f_{max} = \frac{c}{(1200 \text{ nm} - \frac{75 \text{ nm}}{2})} \\ f_{min} = \frac{c}{(1200 \text{ nm} + \frac{75 \text{ nm}}{2})} \end{cases} \Rightarrow \tau = \frac{\mathcal{C}}{\Delta f} = 28.2 \text{ fs}$$

$$\text{Peak Power} = \frac{P_{out}}{f_{rep} \cdot \tau} = \frac{67.5 \text{ mW}}{(1 \text{ kHz})(28.2 \text{ fs})} = 2.4 \text{ GW} < 3.3 \text{ GW}$$

As seen, the additional 7.5 mW obtained with the 1.4 mm- thick BBO crystal do not compensate the broadening in time due to dispersion effects. Therefore, and given that the main problem regarding the observation of sidebands in the two-color photoionization experiment was related to the power of the probe and not to the resolution, the 1 mm- thick BBO crystal was put back in the setup.

To conclude with this section, the measurement of the actual signal pulse duration was acquired using the *frequency-resolved optical gating* (FROG) setup illustrated in **Figure 3.14**. The FROG technique was first developed by Rick Trebino's research group at Los Álamos National Laboratory in the nineties [43], and it is nowadays used for a complete characterization of ultrashort pulses: pulse duration as well as full time-dependent electric field including the optical phase [44]. Although an exhaustive description of this method goes beyond the scope of this work, its principle of operation is similar to the one of an autocorrelator. After technological improvements made possible the generation of sub-picosecond ($< 10^{-12} \text{ s}$) pulses, electronic devices were

no longer suitable for measuring such short durations because they were well below their temporal resolution, typically in the nanosecond range. Since there are very few phenomena as fast as femtosecond pulses, the solution to this issue is measuring a light pulse, indeed, with another light pulse (*correlation*); or more specifically, with itself (*autocorrelation*). The main idea is splitting the light pulse which is to be measured in two replicas and sending each of them to a different arm on an interferometer. The arm length difference and the relative timing of the pulses is adjusted with a piezoelectric delay stage. After recombination, they are both focused into a crystal with a $\chi^{(2)}$ nonlinearity (in this case, a type I, 10 μm –thick BBO). Assuming spatial overlapping, if both pulses are temporally synchronized (total overlap) or the relative time delay is lower than their duration (partial overlap), a second-order effect occurs, which leads to an output of a shorter wavelength that can be detected. By scanning the range for which this second-order effect is detected, the pulse duration can be estimated.

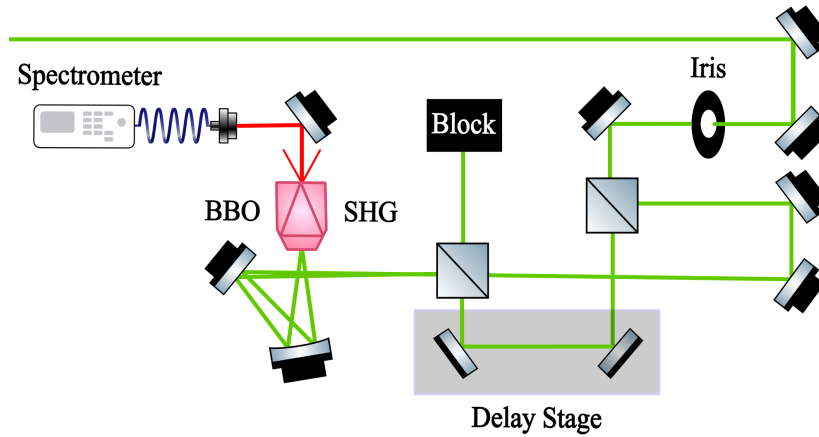


Figure 3.14: FROG setup used to measure the actual pulse duration of the NOPA output.

In our setup, this second-order effect was second harmonic generation. By focusing the two non-collinear replicas in the BBO crystal, three beams at double the fundamental frequency (i.e. at $2 \cdot c/1200 \text{ nm}$) were obtained as output. As sketched in **Figure 3.15** (left), the top and bottom beams are the result of the combination of two “fundamental photons” coming from pulses “b” and “a”, respectively, to give a single photon of twice the fundamental energy. The middle output beam is in turn due to the combination of one fundamental photon from pulse “a” and another fundamental photon from pulse “b” (or viceversa), to give again a doubled-frequency photon, but because of momentum conservation, is emitted along the bisector of the former. Given that both pulses contribute equally to this signal, it will only be visible as long they are temporally overlapped. In other words, the delay range for which the “a + b” signal is visible depends on the duration of the pulse which is to be measured.

Figure 3.15 (right) shows the final result for the actual pulse duration of the NOPA amplified signal, which was calculated using a FROG algorithm³. It has a value of 63.2 fs measured at FWHM, i.e. it is 44.6 fs longer than its Fourier limit. This means

³The implementation of this algorithm and the corresponding figure have not been done by the author of this thesis, they are being shown for completeness.

that the instantaneous frequency

$$\nu(t) = \frac{1}{2\pi} \frac{d\varphi}{dt} \quad (3.6)$$

defined as the temporal derivative of the oscillation phase [45], varies with time due to the effects of chromatic dispersion and nonlinearities (e.g. self-phase modulation). The pulse is said to be *chirped*. This agrees with [3]: “Typically, for visible/near-IR wavelengths, the OPA pulses are positively chirped due to material dispersion introduced by the seed generation process (e.g. by linear propagation of the white light in the sapphire/YAG plate), by optical elements in the signal path (lenses, beam splitters, spectral filters ...) and by the OPA crystals”.

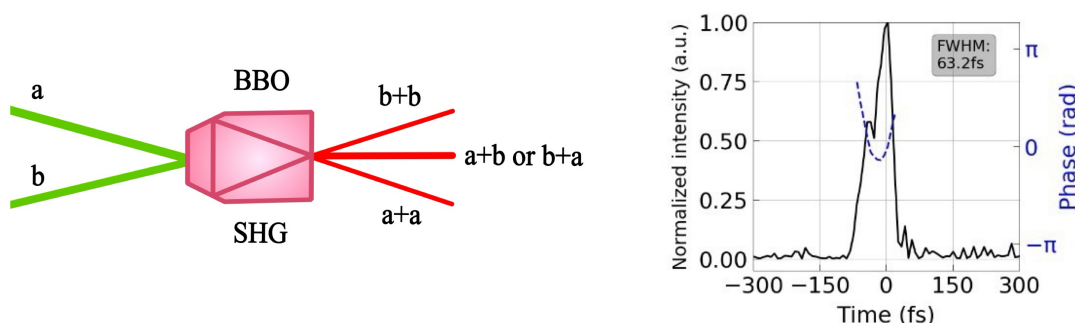


Figure 3.15: Qualitative illustration of the working principle of FROG (left) and measurement of the actual pulse duration of the amplified signal pulse (right).

Connecting with the discussion starting in page 50 about the peak power per pulse and the observation of sidebands, it is worth to consider **Tables 3.4** and **3.5** from the two-color field experiment. Apparently, the probe beam power after the center-hole mirror required to observe sidebands and avoid multiphoton absorption processes must be between 31 mW and 60 mW for ≈ 30 fs pulses, which corresponds to peak powers per pulse between 1 GW and 2 GW. Since the NOPA pulses have twice that pulse duration, an output power of 60 mW measured after the second amplification stage will mean ≈ 1 GW of peak power per pulse before entering the attosecond interferometer. If one considers possible sources of energy loss such as the center-hole mirror itself, where the beam loses $\approx 15\%$ of its energy⁴, the observation of sidebands will not be feasible at this power most likely. Instead, output powers after the second amplification stage close to 100 mW should enable two-color photoionization. Work documented prior to the start of this thesis confirms how values close to this one were once reached. In any case, alternative solutions could be building a third amplification stage or trying to compress the pulse closer to its Fourier limit, which is typically done by compensating the positive group delay dispersion (GDD) of the NOPA pulses with negative one. There exist various optical systems providing negative dispersion in the visible/NIR, like grating pairs, prism pairs, chirped mirrors and adaptive optical systems. According to [3], the best option would be a pair of chirped mirrors or pulse shapers, since they can be designed to correct simultaneously the GDD and other third-order dispersion effects. Nevertheless, they are also expensive and could introduce additional energy losses by absorption.

⁴This value depends on the beam size at the center-hole mirror; the smaller it is, the more energy is lost through the hole. However, the diameter of the output of the Ti:Sa beam measured after the auxiliary telescope is close to the one of the NOPA (≈ 5 mm).

3.3.2 Intensity stability

In RABBITT, the intensity of the sidebands oscillate as a function of the delay between the NIR and XUV fields because of the quantum interference term in Eq.(1.42). Nevertheless, if the intensity of any of the fields fluctuates as a function of time, additional correction terms would be necessary in order to accurately describe the intensity oscillation of the sidebands. This section explains how the intensity stability of the NOPA beam was measured and characterized with the purpose of using it as the probe beam in future RABBITT experiments.

At a fixed power of 3.54 W of the Ti:Sa laser system, the intensity of 999 individual NOPA shots was monitored using a photodiode⁵, in turn connected to an oscilloscope⁶. The acquisition time (t_{aq}) was ca. 4 minutes, with a sampling rate of 1/250 milliseconds. Given the repetition rate of the pump, 1 kHz, this is equivalent to say that 1 in every 250 pulses were analysed.

Figure 3.16 shows a test measurement of the voltage measured by the oscilloscope as a function of time for two different light pulses. The sudden spiking in the nanosecond range, as well as the subsequent exponential decay can be seen. Some small fluctuations are found at the end of both spectra. Although both measurements look indeed very similar, a slightly different peak value is noted if compared closely.

An analysis of the fluctuations of the peak intensity was done first. For every file, the peak value and the corresponding time at which it took place were saved. The following observations were made:

1. The temporal resolution of the measurement was of 0.2 ns.
2. The maximum registered voltage was 186 mV, and the minimum, 168 mV, which means a total fluctuation of less than 10%. The mean registered voltage was 177 mV.

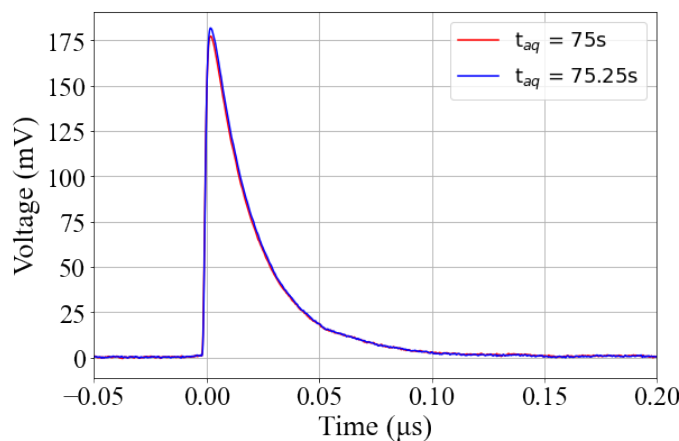


Figure 3.16: Voltage measured by the oscilloscope as a function of time for two different light pulses. In red color, the one corresponding to $t_{aq} = 75$ s, and in blue color, the one measured immediately after the latter, i.e. at $t_{aq} = 75.25$ s.

⁵Model PBM42, from ThorLabs.

⁶Teledyne Lecroy WaveSurfer 4054HD, with serial number LCRY4903C18563.

Figure 3.17 shows the peak-to-peak distribution as a function of acquisition time. First, a blue, dashed line was plotted at the mean peak value (177.5 mV). With the purpose of observing the evolution of the dataset with respect to this value, a linear regression was performed according to the formula $y = ax + b$, where $y \equiv \text{Voltage}$, $x \equiv \text{Acquisition time}$, $a = (6.8 \pm 9.8) \times 10^{-10} \text{ mV/s}$ and $b = (177.4 \pm 0.1) \text{ mV}$. The “scikit-learn” library from Python was used.

In the ideal case, one would expect the slope to be zero and the intercept to be equal to the mean, which would imply that the intensity peaks neither increase or decrease with time, i.e. they stay constant around the mean value. As seen, the value calculated for a is smaller than its associated uncertainty, which shows how the slope is in practice indistinguishable from zero. In turn, the intercept b includes the mean if considered within its uncertainty range. This confirms that the peak intensity of the pulses does not increase linearly over periods of minutes, suggesting stability. This statement was verified when plotting the corresponding histogram, which shows a normal distribution (symmetric by definition) centered at the mean peak value. This is why in order to quantify the total fluctuation of the data set around the mean, the relative standard deviation (RSD)⁷ was used:

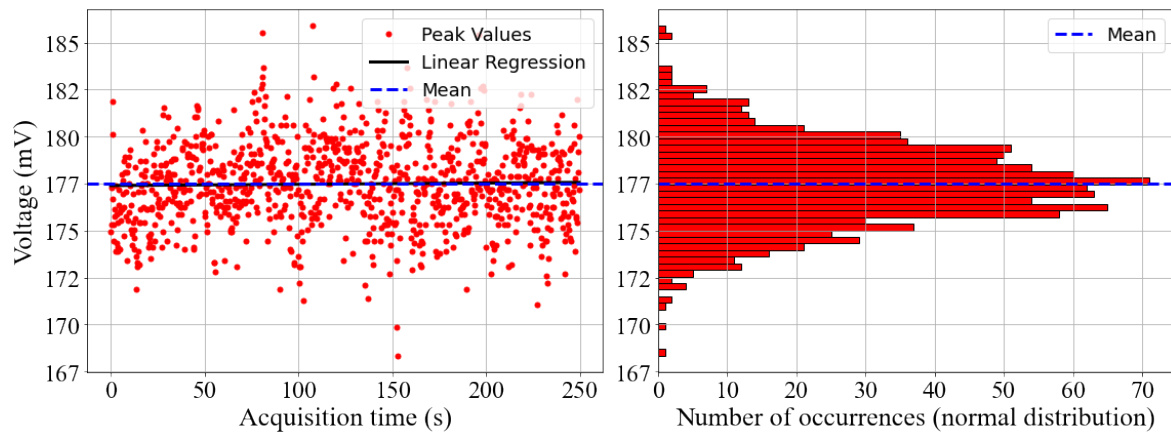


Figure 3.17: Peak-to-peak distribution as a function of acquisition time (left), and normal distribution of the total number of occurrences (right).

$$\text{RSD} = \frac{\sigma}{\mu} = \frac{\sqrt{\frac{\sum_i (x_i - \mu)^2}{N}}}{\mu} = 1.26\% \quad (3.7)$$

where σ is the standard deviation, μ is the mean peak value and x_i are the individual peak values of the data set. The peak intensity from pulse to pulse can thus be considered very stable in concordance with the statement made in section **Practical generation of white light**: when pumped with stable Ti:Sa femtosecond pulses, the white light generated in sapphire crystals has high-quality beam profiles and low pulse-to-pulse fluctuations.

Subsequently, a similar analysis was made but quantifying in this case the area under the curve from $t = -0.05 \mu\text{s}$ to $t = 0.2 \mu\text{s}$ (see **Figure 3.16**). All the voltage values

⁷Also known as the *coefficient of variation* (CV), the *normalized root-mean-square deviation* (NRMSD) or *percent RMS*.

were added in this time interval, and analogously to the previous case, a blue, horizontal line was plotted at the mean integral. A linear regression according to the equation $y = ax + b$, where $y \equiv \text{Integral}$, $x \equiv \text{Acquisition time}$, $a = (2.06 \pm 1.33) \times 10^{-7} \text{ s}^{-1}$ and $b = (22.17 \pm 0.02)$ was performed to see the evolution of the data set with respect of the mean, which was in this case equal to 22.20 (a.u.).

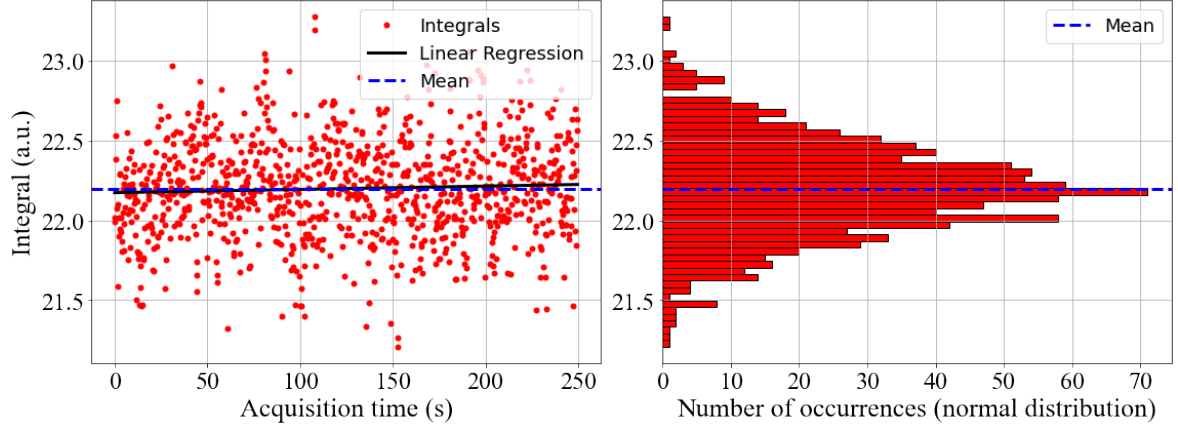


Figure 3.18: Distribution of integrals as a function of acquisition time (left), and normal distribution of the total number of occurrences (right).

In this case, a slightly tendency towards greater values is observed: the slope is still above zero even considering its uncertainty and the intercept does not include the mean, neither. However, the difference is very small, and given that the intensity peaks have been proved to stay stable, this might be due to a slight worse response of the photodiode after some acquisition time. A RSD of 1.36% was estimated in this case, which is again lower than 1.4%. The intensity stability of the NOPA output can thereby be considered stable at least over periods of time in the order of minutes. However, to be completely sure about its suitability for two-color photoionization experiments it would be necessary to measure the intensity over longer acquisition times. Particularly, over hours, since this is often the time it takes to complete this kind of experiments.

3.3.3 CEP stability

The Ti:Sa laser source used in this thesis comes with an active CEP stabilization system. Since the repetition rate of this and most femtosecond oscillators is on the order of 80 MHz [8], it is rather difficult to lock all the CEPs of all the pulses to the same value. Instead, what is typically done is locking the CEP phase change rate, defined as $\Delta\varphi_{\text{CEP}}/T_{rt}$, where T_{rt} is the reciprocal of round trip time in the cavity. This change rate is directly related to the repetition rate of the source, f_{rep} . Pulses with identical CEP are then selected by a pulse picker and sent to the chirped pulse amplifier. Nevertheless, CEP shifts like the one shown in Eq.(1.23) can be introduced from pulse to pulse by the components in the CPA amplifier, the subsequent spectral broadening and pulse compression stages.

The principle of CEP stabilization is better explained in terms of frequency combs. Great part of the formalism presented in the following is based on chapter 3 of [8].

Carrier-Envelope Offset Frequency

When the CEP change rate is kept constant, the electric field of the transformed-limited pulse train at a given point can be expressed as follows⁸:

$$E_{pt}(t) = \sum_{j=-\infty}^{j=\infty} A(t - jT_{rt}) e^{i[\omega_0(t-jT_{rt}) + j\Delta\varphi_{\text{CEP}} + \varphi_{\text{CEP},0}]}, \quad (3.8)$$

where j is the label of a particular pulse in the train, $\varphi_{\text{CEP},0}$ is the CEP of the pulse corresponding to $j = 0$, $A(t)$ is the envelope function of a single pulse in the train and ω_0 is the carrier angular frequency of the pulse. Assuming that the shape and width do not change from pulse to pulse, the laser field in the frequency domain can be obtained applying the Fourier transform to Eq.(3.8):

$$\tilde{E}_{pt}(\omega) = e^{i\varphi_{\text{CEP},0}} \tilde{E}_{sp}(\omega - \omega_0) \sum_{q=-\infty}^{q=\infty} \delta(\omega T_{rt} - \Delta\varphi_{\text{CEP}} - 2\pi q), \quad (3.9)$$

where

$$\tilde{E}_{sp}(\omega) = \int_{-\infty}^{+\infty} E(t) e^{-i\omega t} dt$$

is the electric field of a single pulse in the train in the frequency domain and q is an integer. Eq.(3.9) is the equation of a frequency comb, where the angular frequency of the q th comb tooth is given by

$$\omega_q = 2\pi \frac{q}{T_{rt}} + \frac{\Delta\varphi_{\text{CEP}}}{T_{rt}} \quad (3.10)$$

In terms of frequency,

$$f_q = qf_{rep} + f_0, \quad (3.11)$$

where

$$f_0 = \frac{1}{2\pi} \frac{\Delta\varphi_{\text{CEP}}}{T_{rt}} \quad (3.12)$$

is the so-called *carrier-envelope offset* (CEO) frequency, and it is directly proportional to the CEP phase change $\Delta\varphi_{\text{CEP}}$.

Two frequency combs are represented in **Figure 3.19** (a):

1. In dashed lines, the longitudinal modes of a cavity with no dispersion, i.e. when the group and phase velocities are equal.
2. In solid lines, the one corresponding to the amplified laser pulse in the cavity (that is, considering dispersion effects). Due to the limited bandwidth of the pulse, the number of teeth under the power spectrum is also finite.

The spacing between two adjacent teeth in both cases is the same, and corresponds to the repetition rate of the laser system, f_{rep} . Taking the dashed-line-frequency comb as the reference, it is seen that there exists a certain frequency offset f_0 between both of them. If $\Delta\varphi_{\text{CEP}}$ varies with time, so does f_0 (see Eq.(3.12)).

⁸Note however that the signal output of the NOPA is not Fourier-limited, as seen in **section 3.1.1**. This is therefore an approximation.

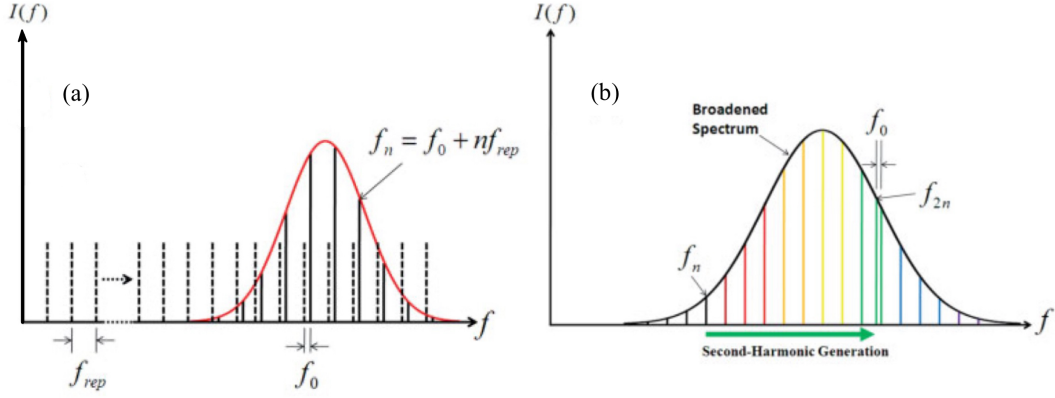


Figure 3.19: (a): frequency comb. The dashed lines represent the modes of the laser when the phase and group velocities of the pulse are equal. The solid lines represent the shifted modes of the laser underneath the gain spectrum in the presence of dispersion. (b): f-to-2f self-referencing technique. f_n is frequency doubled, and f_0 is the difference between f_n and f_{2n} . Taken from [46].

Single Shot f-to-2f Interferometry

What is done in practice is measuring f_0 rather than $\Delta\varphi_{\text{CEP}}$, which are related by Eq.(3.12). When the spectrum of a pulse train covers an octave (i.e. it is extended over a wide range of frequencies, at least until its second harmonic), a low-frequency tooth $f_q = qf_{\text{rep}} + f_0$ can be frequency-doubled to $2f_q = 2(qf_{\text{rep}} + f_0)$ via second harmonic generation, interfering with a high-frequency tooth $f_{2q} = 2qf_{\text{rep}} + f_0$ from the original spectrum at that frequency. The beat frequency from the interference is

$$2f_q - f_{2q} = 2(qf_{\text{rep}} + f_0) - (2qf_{\text{rep}} + f_0) = f_0, \quad (3.13)$$

as illustrated in **Figure 3.19** (b). Therefore, one can quantify how the CEO fluctuates with time and take this as a signal error. By using feedback control techniques, it is possible to stabilize the CEP change rate. This is exactly how the Ti:Sa laser system generates CEP-stable laser pulses.

The goal in this section was to measure the CEP stability of the NOPA output, which although it is seeded by CEP-stable laser pulses, it might not be as stable due to pump power and/or temperature fluctuations. In order to do so, the f-to-2f interferometer sketched in the figure below was built:

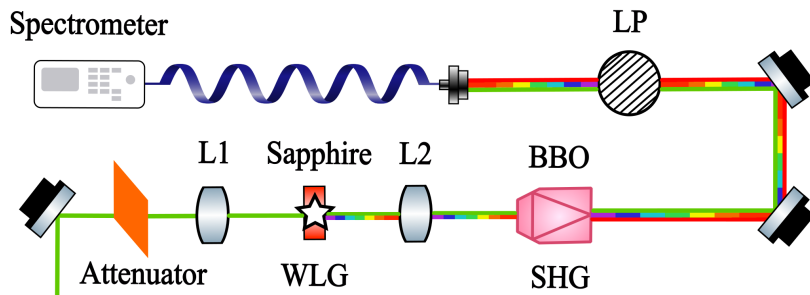


Figure 3.20: Sketch of the f-to-2f interferometer used to measure the CEP stability.

As seen in **Figure 3.12**, the width of the NOPA output extends from around 1125 nm to 1250 nm, which is far from covering an octave. In order to broaden the spectrum, the laser beam at fundamental frequency $f = c/1200$ nm is focused in a 3-mm sapphire plate. Since the laser peak power is higher than the critical power of the sapphire plate, a single filament is formed inside the crystal and the pulses undergo self-phase modulation. White-light generation takes place, and the bandwidth of the now broadened spectrum goes from about 500 nm to 1900 nm, covering an octave. This white light is then focused into a type I, 3 μ m-thick BBO crystal, which converts the 1200 nm light into ≈ 600 nm via second-harmonic generation. The polarization of the second harmonic pulse is perpendicular to that of the 600 nm “original” white light due to the type I phase matching in the BBO (see **Table 1.1**). Then, a polarizer is used to select the common polarization for them to interfere which maximizes the contrast of the fringes. Finally, the interference pattern is recorded by a spectrometer. There is a spectrum phase difference between the two ≈ 600 nm pulses due to dispersion effects in the BBO crystal and in the polarizer. Hence, the final spectrum experiences modulations with a period of τ_g , where τ_g is the time delay caused by group velocity dispersion.

Once the setup was optimized and covered with lids with the purpose of minimizing air fluctuations and vibrations, a 5000 s long (≈ 83 minutes) scan measurement was taken, where the interference spectrum was recorded with a sample rate of 1/500 ms. When the CEP stabilization system of the Ti:Sa laser source was turned off, no interference pattern was observed in the spectrometer because fluctuations on the CEO of the Ti:Sa laser pulses which fed the NOPA caused the fringes to move around randomly. After averaging, intensity peaks were completely washed out. In contrast, when it was turned on interference fringes were clearly visible. As illustrated in **Figure 3.21**, their peaks still match even after averaging, which already suggests a certain degree of stability. Nevertheless, it was also noted that the intensity of the interference peaks decreases slightly after averaging, i.e. there exist indeed some fluctuations. Both of these observations are in agreement with the heatmaps shown in **Figure 3.22**. The intensity of each wavelength is plotted against acquisition time. The horizontal-line pattern indicates that the intensity signal at each wavelength remains approximately constant, which is equivalent to say that *peaks remain locked in position*. Some fluctuations are visible throughout the whole measurement, specially at acquisition times below 1000 s.

Fourier Transform Spectral Interferometry (FTSI)

In the frequency domain, the electric fields of the f and $2f$ pulses can be expressed as follows

$$\tilde{E}_{WL}(\omega) \propto \sqrt{I_{WL}(\omega)} e^{i[\varphi_{WL}(\omega) + \varphi_{\text{CEP}}]}, \quad (3.14)$$

$$\tilde{E}_{SH}(\omega) \propto \sqrt{I_{SH}(\omega)} e^{i[\varphi_{SH}(\omega) + 2\varphi_{\text{CEP}} + \omega\tau_g]}, \quad (3.15)$$

where $I_{WL}(\omega)$ and $I_{SH}(\omega)$ are the intensities of the 600 nm pulse from the white light directly and the second harmonic of the 1200 nm, respectively. $\varphi_{WL}(\omega)$ and $\varphi_{SH}(\omega)$ are their spectral phases. Analogously to Eq.(1.42), a lineout of the interference pattern between the f and $2f$ pulses can be expressed as

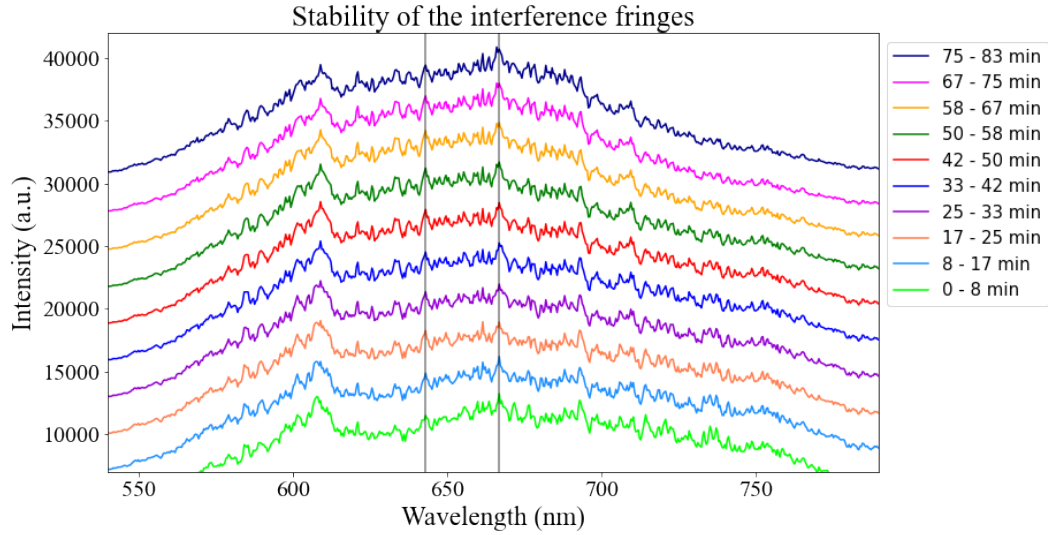


Figure 3.21: Stability of the interference fringes over ≈ 83 minutes. The average of the all the spectra recorded approximately every 8 minutes is represented as a function of the wavelength. An intensity offset has been added to each of them so that they could be visualized one on top of each other. Two vertical, black lines have been plotted to illustrate how the peaks of the interference fringes match.

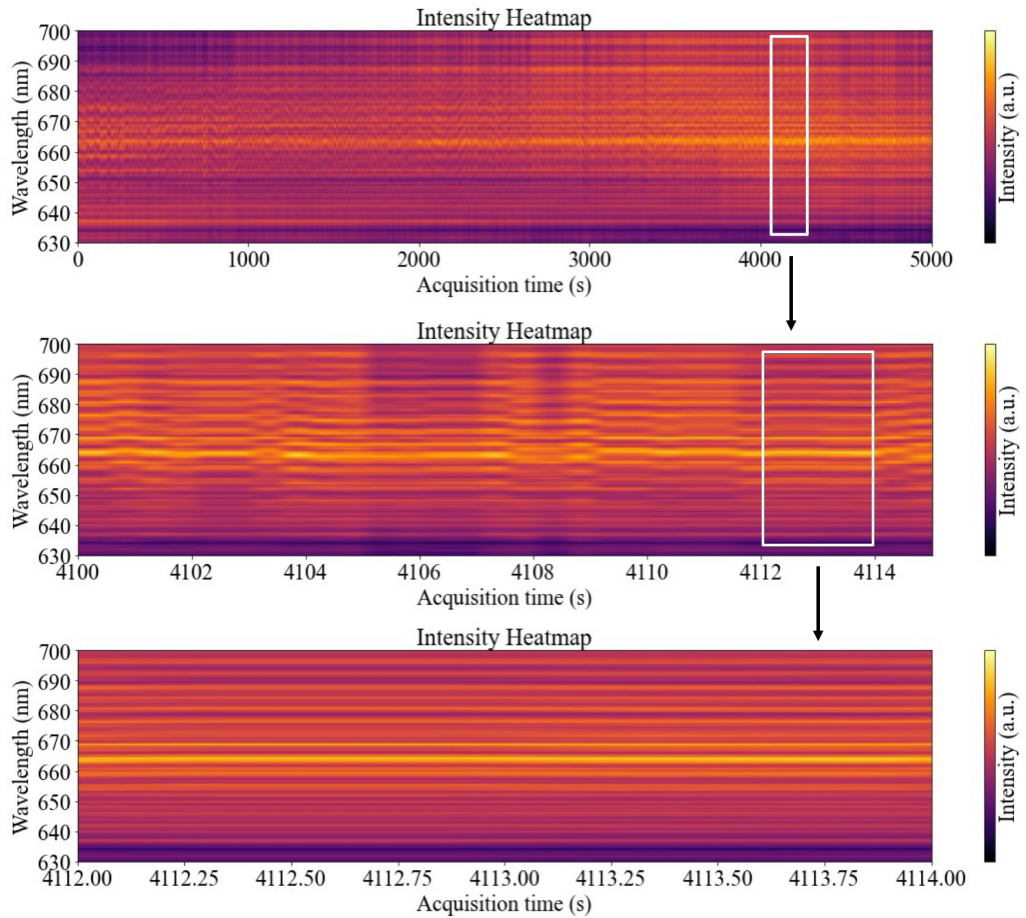


Figure 3.22: Intensity of each wavelength as a function of acquisition time. Shorter time intervals show better stability, as one could expect.

$$\begin{aligned}
I(\omega) &\propto |\tilde{E}_{WL}(\omega) + \tilde{E}_{SH}(\omega)|^2 \\
&= I_{WL}(\omega) + I_{SH}(\omega) \\
&\quad + 2\sqrt{I_{WL}(\omega)I_{SH}(\omega)} \cos(\omega\tau + \varphi_{SH}(\omega) - \varphi_{WL}(\omega) + \varphi_{\text{CEP}}),
\end{aligned} \tag{3.16}$$

According to [47], “it is reasonable to assume that $\omega\tau + \varphi_{SH}(\omega) - \varphi_{WL}(\omega)$ stays constant for one particular frequency component, and thus the phase of $I(\omega)$ can be retrieved as the CEP plus a constant offset”:

$$I(\omega) \propto I_{WL}(\omega) + I_{SH}(\omega) + 2\sqrt{I_{WL}(\omega)I_{SH}(\omega)} \cos(\mathcal{C} + \varphi_{\text{CEP}}) \tag{3.17}$$

Given that the seed pulse used in the NOPA is obtained via WLG starting from the fundamental frequency-pulse, the CEP of the driver is transferred to the seed, and

$$\varphi_{\text{CEP, signal}} = \varphi_{\text{CEP, Ti:Sa}} - \pi/2 + c_1, \tag{3.18}$$

where c_1 is a constant representing the accumulated phase due to propagation [3]. Given that $\varphi_{\text{CEP, Ti:Sa}}$ is constant because of the stabilization system of Ti:Sa laser source, and since all the other terms are also constants, $\varphi_{\text{CEP, signal}}$ should in principle be constant as well.

In order to extract φ_{CEP} from the interference term in Eq.(3.17), the algorithm “Fourier Transform Spectral Interferometry” (FTSI) was implemented. From each spectrum, only the wavelengths for which the interference fringes were more obvious were considered, i.e. from 640 nm to 680 nm. After normalizing the intensities, they were plotted as function of frequency instead of wavelength, keeping in mind that from energy conservation

$$I(\lambda)d\lambda = I(f)df, \quad \text{where } f = \frac{c}{\lambda} \quad \Rightarrow \quad I(f) = I(\lambda)\frac{d\lambda}{df} = I(\lambda) \cdot \frac{c}{f^2} \tag{3.19}$$

Then, the frequency vector was equispaced⁹ and the values of $I(f)$ were interpolated to this new frequency vector using the function “interp1d()” from the “scipy” library of Python. A Super Gaussian filter was applied next with the purpose of reducing low-frequency noise, according to the expression

$$e^{-\left(\frac{f-f_0}{\Delta f}\right)^\alpha} \tag{3.20}$$

Here, f is the vector of equispaced frequencies, f_0 is the central frequency, which was calculated as the mean of the minimum ($c/680$ nm) and maximum ($c/640$ nm) considered frequencies; Δf is the frequency width, set to 7 THz; and $\alpha = 8$.

The electric field was calculated taking the square root of the interpolated and filtered spectrum. A Fourier transform was applied subsequently to express it in the time domain using the function “np.fft.fft()” from the NumPy library, and after this, intensities as a function of time were obtained by modulus squaring the electric field. The relative CEP was extracted as the phase of the electric field at the first, non-direct current intensity peak in the time domain. For this, the NumPy function “np.angle()” was implemented.

⁹This step is necessary in order to apply the Fast Fourier Transform (FFT).

Figure 3.23 summarizes the followed procedure:

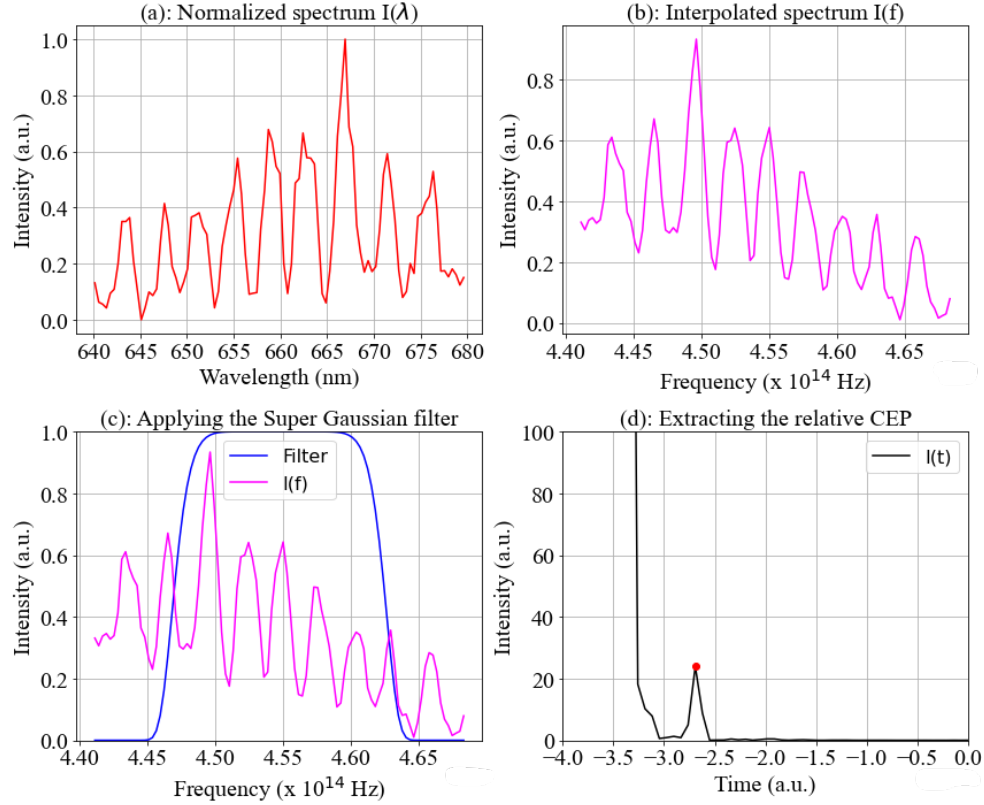


Figure 3.23: (a): selection of the interference fringes, (b): interpolated spectrum over the equispaced frequency vector, (c): application of the Super Gaussian filter, (d): intensities as a function of time and first non-dc peak, from which the relative CEP was extracted.

Finally, phases were plotted as a function of acquisition time:

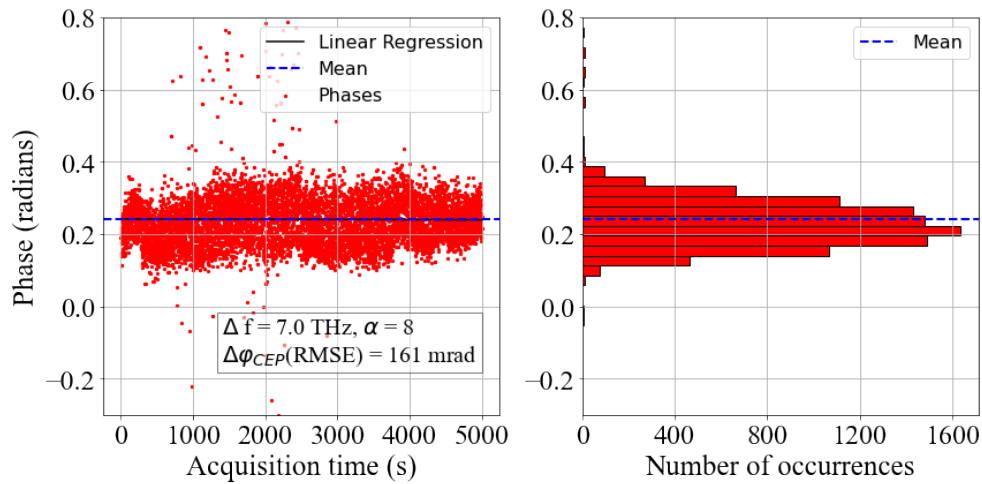


Figure 3.24: Relative phases as a function of time. A RMSE of 161 mrad was calculated for the relative CEP when a frequency width of $\Delta f = 7$ THz and an exponent $\alpha = 8$ were set in the Super Gaussian filter. The calculated phases follow a normal distribution centered approximately around the mean value.

A linear regression was performed using the “scikit-learn” library, according to the formula $y = ax + b$, where $y \equiv \text{Phase}$, $x \equiv \text{Acquisition time}$, $a = (-0.54 \pm 1.11) \times 10^{-3} \text{ mrad/s}$ and $b = (242 \pm 3) \text{ mrad}$. Analogously to the previous section, where the intensity stability was analysed, in the ideal case one would expect the slope to be zero and the intercept to be equal to the mean, which in this case was 241 mrad. This would be translated into a stable evolution of the dataset. As seen, the values for a and b calculated with the linear fit include the zero and the mean, respectively, if considered within their uncertainty range, suggesting CEP stability over a period of ≈ 1.5 hours.

The root mean square error was calculated, according to

$$\text{RMSE} = \sqrt{\frac{\sum_i^N (x_i - \hat{x}_i)^2}{N}}, \quad (3.21)$$

where x_i are the obtained phases, $\hat{x}_i$ are their predictions based on the linear regression model, and N is the number of acquired phases (one for each spectrum). This parameter indicates how spread are the measured values with respect to the fit, and was chosen to make comparisons with the values found in the literature.

As seen in **Figure 3.22**, the apparently most stable interval starts after $t_{aq} \approx 3000 \text{ s}$. If only those spectra are considered, $\Delta\varphi_{CEP}(\text{RMSE})$ decreased down to 58 mrad.

Comparing with the values found in the literature [8] for stabilized CEP drifts, which are on the order of $\approx 100 \text{ mrad}$, the NOPA pulses can thereby be considered CEP stable over periods of time of minutes, in agreement with the hypothesis stated under Eq.(3.18). Nevertheless, three important remarks should be made regarding this aspect:

1. The calculation of $\Delta\varphi_{CEP}(\text{RMSE})$ is highly sensitive to the frequency width and to the exponent of the Super Gaussian filter. Given that this filter is applied to reduce low-frequency noise, these parameters have direct effects on the position of the first non-dc intensity peak, and thus, on the calculation of the relative CEP. Variations of a few hundred of miliradians were observed when varying the width over 1 THz. For the same width Δf as in **Figure 3.24** and for $\alpha = 10$, $\Delta\varphi_{CEP}(\text{RMSE})$ gives 67 mrad.
2. The CEP stability depends strongly on the pump power. In case of being unstable due to temperature changes and consequent misalignment of the Ti:Sa laser system during the day, the CEP stability could be compromised. The day these measurements were taken, the Ti:Sa output power was set to $\approx 3.5 \text{ W}$ (in order to not exceed the damage threshold of the BBO crystals, an additional 50/50 beam splitter before the first stage was needed).
3. If in the future a prism-based compressor is used to compress the NOPA pulses to obtain greater peak energies per pulse, it should be noted that the CEP depends strongly on the separation between prisms.
4. CEP shifts can be also introduced in dispersive media where $v_g \neq v_\varphi$. This is the case for fused silica, which is often used as a window material for vacuum chambers in attosecond pulse generation setups (although as long as $\Delta\varphi_{CEP}$ remains constant, this in principle would not be a critical problem).

Chapter 4

Summary and Outlook

Summary

This thesis has been focused on the optimization of an attosecond interferometer which enables the realization of two-color photoionization experiments. Additionally, the output of an optical parametric amplifier (OPA) has been characterized with the purpose of using it in further two color-field photoionization experiments.

On the first place, a one color-field experiment was performed. The near-infrared (NIR) femtosecond output of a commercially produced Titanium-doped Sapphire laser system was focused on a gas target of argon, creating an attosecond pulse train in the XUV range via HHG. The resulting harmonic spectrum was recorded using an XUV spectrometer placed at the end of the beamline, and it consisted of 9 odd harmonics separated by twice the energy of the driving field ($E_{NIR} = \frac{hc}{\lambda_{NIR}} \approx 1.59$ eV). Because of the aluminum filter employed to block the residual NIR radiation, all harmonics below the 11th were not detected. Additionally, the efficiency of the reflection grating in the spectrometer was too low to transmit harmonic 11th, so the peaks appearing in the spectrum ranged from the 13th to the 29th, inclusive.

This XUV radiation was then focused on an a second gas target of argon, with $I_p^{Ar} = 15.76$ eV, where ionization took place. The kinetic energies of the resulting photoelectrons were measured with a VMI spectrometer (placed in the beamline before the XUV spectrometer), and the six visible rings of increasing radii were associated to harmonics 11th, 13th, 15th, 17th, 19th and 21st, respectively. Raw images were Abel-inverted using the pBASEX-Method, and the photoelectron spectrum was extracted. The relative intensity of the harmonic peaks was discussed keeping in mind the atomic absorption cross section of argon atoms as a function of energy. Some possible broadening mechanisms were also described.

Then, a two color-field experiment was performed, where the probe beam in the attosecond interferometer was unblocked and recombined with the XUV radiation described in the paragraph above. It was realized that the probe beam lost around 80% of its energy in the interferometer due to its great beam diameter. At a measured power of ≈ 31 mW after the center-hole mirror, sidebands were not visible. In order to solve this issue an auxiliar telescope was built and installed before the attosecond interferometer. The reduction of its diameter to about 1/2 of its original value led to a significant improvement on the energy loss, now close to 50%. The measured power after the center-hole

mirror was now ≈ 60 mW. As a result of photoelectron interaction with the NIR field, sidebands were finally visible in the VMI images halfway through the rings associated to ionization by harmonics. The extraction of the photoelectron spectrum from the Abel-inverted image showed their high relative intensity, which suggested the existence of multiphoton absorption processes due to the now too-high optical intensity of the probe beam. This assumption was supported by the observation of a hot spot in the origin of the VMI rings.

The characterization of the output of a home built non-collinear OPA, planned to be used as the probe beam in future two color-field experiments, was regarded on the last place. A final power of (60 ± 3) mW was reached after the second amplification stage for the signal, centered at $\lambda \approx 1200$ nm and corresponding to a Fourier limit of 18.6 fs. The use of a thicker crystal increased the output power by about ≈ 7.5 mW. However, the asymmetry of its spectrum was accentuated, it did not peak at exactly $\lambda = 1200$ nm and the gain in power did not compensate for the broadening of the pulse duration due to dispersion. Therefore, the thinner crystal was decided to be more appropriate for the setup. The actual pulse duration of the NOPA output under this configuration was measured using a FROG setup, and was estimated to be ≈ 63.2 fs. Given that this is twice the duration of the Ti:Sa output pulses, the peak power of the NOPA pulses results to be half of the latter. Consequently, sidebands may not be visible when the NOPA beam is employed in the future for two-color photoionization experiments under the current status. A third amplification stage or a compression of the pulse to its Fourier limit might be necessary to reach suitable peak powers per pulse, estimated to be between 1 GW and 2 GW.

Next, the intensity of the NOPA output pulses was proved to be stable over periods of time of minutes. The intensity of 999 individual shots was monitored over 4 minutes, with a sampling rate of 1/250 milliseconds. The analysis of their peak values showed a constant and stable evolution of the dataset, and a relative standard deviation below 1.4% demonstrated a very low fluctuation around the mean value.

To conclude, the CEP stability was characterized. Given that the Ti:Sa laser system has an active CEP stabilization system and since the seed pulse in the NOPA is obtained via white light generation starting from the fundamental-frequency pulse, the CEP of the signal was expected to be stable. In order to prove it, an f-to-2f interferometer was built. The spectrum of the signal was first broadened via white light generation and frequency-doubled via second harmonic generation. The f and $2f$ components, respectively, interfered at ≈ 1200 nm/2, and the stability of the fringes was monitored over ≈ 83 minutes. In order to measure the relative CEP drift, the Fourier Transform Spectral Interferometry (FTSI) algorithm was implemented. A final value of $\Delta\varphi_{CEP}$ (RMSE) of 161 miliradians was calculated. This result is in agreement with the typical numbers found in the literature for CEP-stabilized drifts [8].

Outlook

Regarding the work done in this thesis, a possible improvement could be the reexamination of the motorized stages inside the HHG Chamber. It was noted experimentally how moving them shifted the alignment slightly, a critical issue for observing sideband oscillations. Besides, significant drops of power of the Ti:Sa laser system were also observed throughout the day. This was due both to temperature fluctuations and to the worsening of the windows in the preamplification and booster stages, which will be changed soon. Finally, an optimization of the LabView programs might be helpful, since they sometimes crashed during the measurements.

In the upcoming weeks, the RABBITT technique could be implemented in order to reconstruct the attosecond pulse train created via HHG in the first place. Then, the NOPA output will be used as the probe beam. Given that it covers a different optical length than the Ti:Sa output before entering the attosecond interferometer, adjustments will have to be done in the HHG chamber in order to accomplish temporal overlap. Before this, it might be beneficial to optimize the peak power per pulse as discussed in **subsection 3.3.1**, either by the implementation of a third amplification stage or by compressing the pulses. Note however that a compression system might also introduce additional absorption losses and could have effects on the CEP of the pulses. Additionally, it would be ideal to replace the auxiliar telescope shown in **Figure 3.7** based on lenses by another one based on focusing mirrors. This way, dispersion effects would be minimized. After recombination with the XUV radiation, and since $\lambda_{probe} \approx 3/2\lambda_{pump}$, not one but two sidebands will appear between each odd harmonic in the two-color photoionization process, as sketched in **Figure 4.1**.

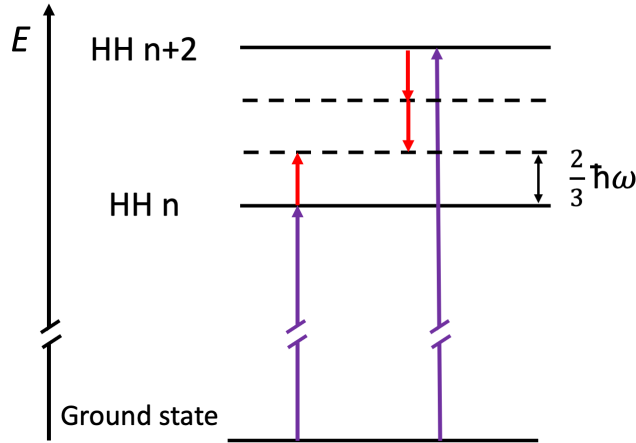


Figure 4.1: Schematic view of the energy levels in the upcoming two-color photoionization experiment, where the energy of the probe field is $2/3$ the energy of the pump field used to create the harmonics via HHG, $E_{probe} = 2/3 E_{pump}$. The two sidebands appearing between consecutive harmonics can each be populated through two paths characterized by a different number of exchanged NIR (i.e. probe) photons (for simplicity, only the population of the lower sideband is shown in the figure): the absorption of one photon of the harmonic n and one (two) NIR photon, or the absorption of one photon of the harmonic $n + 2$ and the emission of two (one) NIR photons.

This project has been part of a bigger one whose final goal is to retrieve information about relative phases in photoionization processes, providing a much more accurate

description of ultrafast electron dynamics. Attosecond time delays in this kind of processes are nowadays studied with two experimental approaches: RABBITT, which uses an attosecond pulse train; and the streaking method, where an isolated attosecond pulse triggers the photoionization process. Both of these techniques superimpose high-frequency photons with a probe field to monitor this process in time, and the interaction of photoelectrons with this probe field adds a certain time delay to the photoionization time delay, which is the observable of interest. In RABBITT, it is often referred to as “continuum-continuum (CC) time delay”, and it has non-negligible effects on the measurement.

Back in Eq.(1.42), it was shown how the phase of an oscillating sideband (due to the time delay between the NIR and XUV fields) can be decomposed in an atomic phase $\Delta\phi_A$ plus the phase difference between consecutive harmonics $\Delta\phi_{XUV}$, i.e. the spectral dispersion of the pulses in the attosecond pulse train (which can be used to estimate the average XUV pulse duration). More precisely,

$$\Delta\phi = \Delta\phi_{XUV} + \underbrace{\Delta\phi_W + \Delta\phi_{CC}}_{\Delta\phi_A}, \quad (4.1)$$

where $\Delta\phi_W$ is related to the so-called “Eisenbud-Wigner-Smith (EWS) time delay”, the observable of interest. $\Delta\phi_{CC}$ is the additional phase shift as a result of photoelectron interaction with the probe field and which leads to the continuum-continuum time delay, contaminating the measurement. At present, it has been estimated mainly with theoretical models, although there exist already measurements schemes to determine it experimentally [48] based on the RABBITT technique. Using the NOPA output as the probe beam in two-color photoionization experiments (i.e. generating two sidebands instead of only one) will enhance the interaction with the NIR field, contributing in a positive way to the search for $\Delta\phi_{CC}$.

Chapter 5

Appendix

5.1 Nonlinear optics

When an electromagnetic wave propagates through a medium, its associated electric field induces a force on the electrons, giving rise to a polarization which can in turn affect the incident wave. This perturbation is negligible at low field intensities; the behaviour is said to be linear in these cases:

$$\vec{P} = \varepsilon_0 \chi \vec{E}, \quad (5.1)$$

where $\vec{P}$ is the polarization, ε_0 is the permittivity of free space, χ is the susceptibility of the dielectric medium and $\vec{E}$ is the electric field. Induced polarization and electric field are thus parallel.

Nevertheless, at high enough intensities, nonlinear effects such as new-frequencies generation, intensity-dependent refractive indexes or ultrashort pulses are observed. Therefore, Eq.(5.1) no longer holds and must be rewritten so that higher-order terms are taken into account:

$$P = \varepsilon_0 [\chi E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \dots] \quad (5.2)$$

As it will be detailed in the upcoming pages, the tensor nature of the susceptibility becomes relevant and each of its orders is responsible of different phenomena.

The physical explanation to all this is that in reality electrons are not bounded to their corresponding nucleons by simple harmonic potentials as the Lorentz Model estimates. At low external field intensities, electrons do not get very far from the equilibrium point, and consequently the harmonic approximation provides an analytical and fairly accurate solution to the equations of motion. However, at high intensities the interaction is such that electrons do get far enough from the equilibrium point and feel the anharmonicity of the potential:

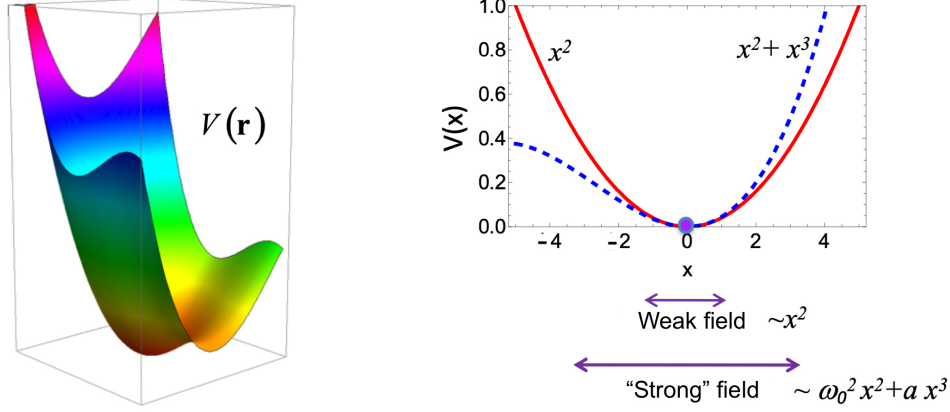


Figure 5.1: More realistic potential and harmonic potential approximation. At great distances from the equilibrium point, anharmonicity becomes relevant. Adapted from [49].

Considering a third-order term in our Taylor expansion of the potential around the minimum, and for the sake of simplicity, a one dimensional problem:

$$V(x) \approx V_0 + \left. \frac{dV}{dx} \right|_{x=0} x + \frac{1}{2} \left. \frac{d^2V}{dx^2} \right|_{x=0} x^2 + \frac{1}{6} \left. \frac{d^3V}{dx^3} \right|_{x=0} x^3 + \dots = \frac{1}{2} m \omega_0^2 x^2 + \frac{1}{3} m A_2 x^3 + \dots, \quad (5.3)$$

where

$$\omega_0^2 = \left. \frac{1}{m} \frac{d^2V}{dx^2} \right|_{x=0} \quad \text{and} \quad A_2 = \left. \frac{1}{2m} \frac{d^3V}{dx^3} \right|_{x=0}$$

The constant term V_0 can be cancelled since the origin of the potential is arbitrary and the first derivative with respect to the position is zero by definition of minimum.

The corresponding bounding force therefore reads

$$F_{bound}(x) = -\nabla V(x) = -m\omega_0^2 x - mA_2 x^2, \quad (5.4)$$

and according to Newton's Second Law, the product of the mass times the acceleration will then be the sum of the involved forces:

$$m \frac{d^2 x}{dt^2} = F_{ext} + F_{bound}, \quad (5.5)$$

where

$$F_{ext} = -eE_x(t) = -e(E_\omega e^{-i\omega t} + E_\omega^* e^{i\omega t})$$

refers to the external force.

From Eq.(5.5), the complete equation of motion is then

$$m \frac{d^2 x}{dt^2} = -e(E_\omega e^{-i\omega t} + E_\omega^* e^{i\omega t}) - m\omega_0^2 x - mA_2 x^2, \quad (5.6)$$

which cannot be analytically solved due to the anharmonic term. Nevertheless, applying a perturbative treatment valid when $A_2 x^2 \ll \omega_0^2 x$, an approximate solution of the time-dependent trajectory of the electron is finally reached:

$$x(t) = x_{\omega}^{(1)}e^{-i\omega t} + x_0^{(2)} + x_{\omega}^{(2)}e^{-i2\omega t} + \dots + \text{c.c.}, \quad (5.7)$$

where the superscripts in parenthesis denote the i th-order solutions. Thus, the electron not only oscillates at the frequency of the external field, but also at twice that frequency. Additionally, it experiences a displacement independent of time: the non-linear behaviour becomes now obvious. In other words, and connecting with what was explained before, the intensity of the incident electric field is such that it makes electrons vibrate in different ways, which cannot be described by a simple harmonic potential.

Analogously for the polarization:

$$P(t) = P_L(t) + P_{NL}(t) = P_{\omega}^{(1)}e^{-i\omega t} + \underbrace{P_0^2 + P_{\omega}^{(2)}e^{-i2\omega t} + \dots}_{P_{NL}(t)} + \text{c.c.}, \quad (5.8)$$

Several nonlinear effects are found depending on the intensity of the driving fields and on the amount of nonlinear terms considered, summarized in the following table:

Table 5.1: First-, second- and third-order nonlinear effects.

1 st Order	2 nd Order	3 rd Order
Linear Dispersion	Optical Rectification Second Harmonic Generation Sum Frequency Generation Difference Frequency Generation Pockels Effect Optical Parametric Amplification	Third Harmonic Generation Four-Wave Mixing Kerr Effect Self-focusing, Self-Steepening & Self-Phase Modulation White Light Continuum Generation

Nevertheless, they do not all show at the same time and under the same conditions. Due to the wave nature of the electron demonstrated by Louis-Victor de Broglie [50], electrons oscillating at a certain frequency can be studied using the Undulatory Theory, and therefore, crucial wave phenomena such as interference must be carefully regarded. In order for a certain nonlinear process to be energetically efficient, a constructive interference between the involved waves must be accomplished. The *phase-mismatch* term Δk describes the exchange of energy between waves along their propagation, and as we shall see in the following sections, efficiency gets maximized when $\Delta k = 0$. The new-frequency generation is a perfectly coherent process; however, waves with different frequencies propagating in the same medium experience different refractive indexes due to dispersion, and therefore propagate at different velocities. Consequently, when they exit this medium (typically a crystal), there is a (most likely) chance that their phases do not match, leading to a destructive interference which will not amplify the signal (see **Figure 5.2**). The solution is to make them propagate at the same velocity. How so, bearing in mind that they are experiencing different refractive indexes? *Making them seeing* the same refractive index so that they propagate at the same velocity, indeed: this is where anisotropic media play a fundamental role.

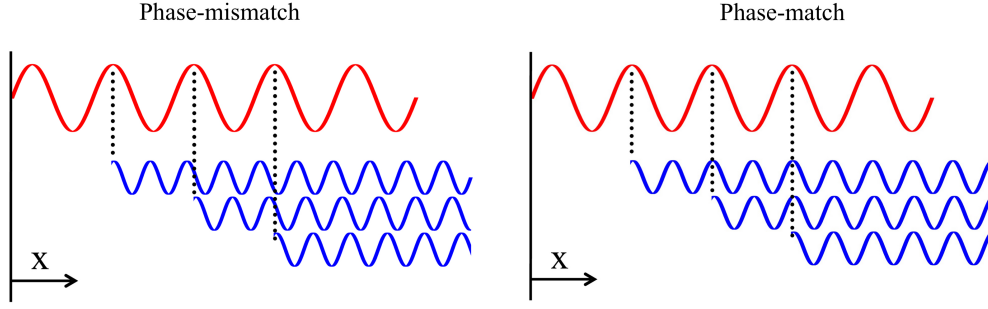


Figure 5.2: Phase-matching of the copropagating waves. Taken from [49].

5.2 Anisotropic media

In Optics, “isotropy” refers to the directional independence of a certain physical property. It is a consequence of the Lorentz Model, where as it was already shown, the induced polarization is parallel and proportional to the incident electric field ¹:

$$\vec{P} \parallel \vec{E} \rightarrow \vec{P}_\omega = \varepsilon_0 \chi(\omega) \vec{E}_\omega,$$

Bearing in mind the relevant effects of the anharmonic potentials in nonlinear optics and therefore the invalidity of the Lorentz Model, “anisotropy” becomes an important quality of some materials, causing them to have a different refractive index in each direction. Given that the optical response of the polarization is no longer linear to the electric field and that ε_0 is a physical constant, the susceptibility must not be treated as a scalar in this case, but as a second-rank tensor:

$$\vec{P} \parallel \vec{E} \rightarrow \vec{P}_\omega = \varepsilon_0 \hat{\chi}(\omega) \vec{E}_\omega \quad \Rightarrow \quad \begin{pmatrix} P_{\omega,x} \\ P_{\omega,y} \\ P_{\omega,z} \end{pmatrix} = \varepsilon_0 \begin{pmatrix} \chi_{11}(\omega) & \chi_{12}(\omega) & \chi_{13}(\omega) \\ \chi_{21}(\omega) & \chi_{22}(\omega) & \chi_{23}(\omega) \\ \chi_{31}(\omega) & \chi_{32}(\omega) & \chi_{33}(\omega) \end{pmatrix} \begin{pmatrix} E_{\omega,x} \\ E_{\omega,y} \\ E_{\omega,z} \end{pmatrix}$$

Analogously for the electric displacement field $\vec{D}$:

$$\vec{D}_\omega = \underbrace{\varepsilon_0(1 + \hat{\chi}(\omega))}_{\hat{\varepsilon}} \vec{E}_\omega = \hat{\varepsilon} \vec{E}_\omega \quad \Rightarrow \quad \begin{pmatrix} D_{\omega,x} \\ D_{\omega,y} \\ D_{\omega,z} \end{pmatrix} = \begin{pmatrix} \varepsilon_{11}(\omega) & \varepsilon_{12}(\omega) & \varepsilon_{13}(\omega) \\ \varepsilon_{21}(\omega) & \varepsilon_{22}(\omega) & \varepsilon_{23}(\omega) \\ \varepsilon_{31}(\omega) & \varepsilon_{32}(\omega) & \varepsilon_{33}(\omega) \end{pmatrix} \begin{pmatrix} E_{\omega,x} \\ E_{\omega,y} \\ E_{\omega,z} \end{pmatrix},$$

where $\hat{\varepsilon}$ is now the dielectric tensor associated with the corresponding medium.

Assuming certain conditions such as transparency, absence of external magnetic fields as well as of optical activity, the dielectric tensor becomes real and symmetric:

$$\varepsilon_{ij}(\omega) = \varepsilon_{ji}(\omega), \quad (5.9)$$

¹Since the material’s response is not strictly instantaneous, it is convenient to work in the spectral domain.

and as a consequence, it will be always possible to find a base which diagonalizes it. Eq. (5.2) will then get reduced to

$$\begin{pmatrix} D_{\omega,x} \\ D_{\omega,y} \\ D_{\omega,z} \end{pmatrix} = \begin{pmatrix} \varepsilon_x(\omega) & 0 & 0 \\ 0 & \varepsilon_y(\omega) & 0 \\ 0 & 0 & \varepsilon_z(\omega) \end{pmatrix} \begin{pmatrix} E_{\omega,x} \\ E_{\omega,y} \\ E_{\omega,z} \end{pmatrix}, \quad (5.10)$$

where $\varepsilon_x(\omega)$, $\varepsilon_y(\omega)$ and $\varepsilon_z(\omega)$ are the so-called *principal axis*. In the most general case, $\varepsilon_x(\omega) \neq \varepsilon_y(\omega) \neq \varepsilon_z(\omega)$. These are the so-called *biaxial media*. While a full mathematical derivation to analyse the behaviour of light in anisotropic media is not the goal of this section, the analysis comes from the Maxwell Equations, which for harmonic waves read:

$$\begin{aligned} \nabla \times \vec{E}_\omega(\vec{r}) &= i\mu_0\omega \vec{H}_\omega(\vec{r}) & \nabla \times \vec{H}_\omega(\vec{r}) &= [-i\omega\hat{\varepsilon}_\omega(\vec{r}) + \hat{\sigma}_\omega(\vec{r})]\vec{E}_\omega(\vec{r}) \\ \nabla \cdot [\hat{\varepsilon}_\omega(\vec{r})\vec{E}_\omega(\vec{r})] &= \rho_\omega(\vec{r}) & \nabla \cdot \vec{H}_\omega(\vec{r}) &= 0, \end{aligned}$$

where $\vec{E}_\omega(\vec{r})$ and $\vec{H}_\omega(\vec{r})$ are the electric and magnetic fields, respectively, μ_0 is the magnetic permeability in vacuum, $\hat{\sigma}_\omega(\vec{r})$ is the conductivity and $\rho_\omega(\vec{r})$ is the charge density. Applying the curl operator to the first equation and combining with the second one, the wave equation is reached.

$$\nabla [\nabla \cdot \vec{E}_\omega(\vec{r})] \nabla^2 \vec{E}_\omega(\vec{r}) = -i\mu_0\omega[\hat{\sigma}_\omega(\vec{r}) - i\omega\hat{\varepsilon}_\omega(\vec{r})]\vec{E}_\omega(\vec{r}) \quad (5.11)$$

Assuming a neutral ($\rho_\omega = 0$), dielectric ($\sigma_\omega = 0$) and homogeneous ($\varepsilon_\omega, \chi_\omega \neq f(\vec{r})$) medium, it gets simplified a bit more:

$$\nabla [\nabla \cdot \vec{E}_\omega(\vec{r})] - \nabla^2 \vec{E}_\omega(\vec{r}) = \mu_0\omega^2\hat{\varepsilon}_\omega\vec{E}_\omega(\vec{r}) \quad (5.12)$$

Subsequently, the wave number $k_0 = \frac{2\pi}{\lambda} = \frac{\omega^2}{c^2} = \mu_0\varepsilon_0\omega^2$, the refractive index $n_\omega = \sqrt{\varepsilon_{rel,\omega}}$ (where $\varepsilon_{rel,\omega} = \varepsilon_{medium,\omega}/\varepsilon_0$ is the relative dielectric constant) and the direction of wave propagation $\vec{s}$ are introduced. Eq.(5.12) gets simplified to

$$n_\omega^2 [\vec{E}_\omega(\vec{r}) - (\vec{s} \cdot \vec{E}_\omega(\vec{r}))\vec{s}] = \frac{\hat{\varepsilon}_\omega}{\varepsilon_0} \vec{E}_\omega(\vec{r}) = \frac{1}{\varepsilon_0} \vec{D}_\omega(\vec{r}) \quad (5.13)$$

Finally, solving by components and using $\nabla \cdot \vec{D} = \rho = 0 \Rightarrow \vec{s} \cdot \vec{D} = 0$, the Fresnel Equation is obtained:

$$\left[\frac{\varepsilon_x}{\varepsilon_x - \varepsilon_0 n^2} s_x^2 + \frac{\varepsilon_y}{\varepsilon_y - \varepsilon_0 n^2} s_y^2 + \frac{\varepsilon_z}{\varepsilon_z - \varepsilon_0 n^2} s_z^2 \right] (\vec{s} \cdot \vec{E}) = 0, \quad (5.14)$$

The first obvious solution is $(\vec{s} \cdot \vec{E}) = 0$, which implies $\varepsilon_x = \varepsilon_y = \varepsilon_z$, i.e., it corresponds to isotropic media. We are therefore interested in finding the solution for

$$\left[\frac{\varepsilon_x}{\varepsilon_x - \varepsilon_0 n^2} s_x^2 + \frac{\varepsilon_y}{\varepsilon_y - \varepsilon_0 n^2} s_y^2 + \frac{\varepsilon_z}{\varepsilon_z - \varepsilon_0 n^2} s_z^2 \right] = 0, \quad (5.15)$$

which can be conveniently written as

$$s_x^2(u_y^2 - u^2)(u_z^2 - u^2) + s_y^2(u_z^2 - u^2)(u_x^2 - u^2) + s_z^2(u_x^2 - u^2)(u_y^2 - u^2) = 0, \quad (5.16)$$

where

$$u = \frac{c}{n} \quad (5.17)$$

is the *phase velocity*.

Optical Parametric Amplification is mostly interested in *uniaxial media*, where $\varepsilon_x(\omega) = \varepsilon_y(\omega) \neq \varepsilon_z(\omega)$. Consequently, from Eq.(5.10), the dielectric tensor gets reduced to

$$\varepsilon(\omega) = \begin{pmatrix} \varepsilon_x(\omega) & 0 & 0 \\ 0 & \varepsilon_x(\omega) & 0 \\ 0 & 0 & \varepsilon_z(\omega) \end{pmatrix}, \quad (5.18)$$

which indicate that there exists a plane without any preferred directions, called *isotropic plane*, and an additional direction with different properties, the *optical axis*, which by convention is usually chosen to be the z-axis. It is possible now to define two refractive indexes characteristic of the material itself:

$$n_o^2 \equiv \frac{\varepsilon_x}{\varepsilon_0} = \frac{\varepsilon_y}{\varepsilon_0} = \frac{c^2}{u_o^2} \quad \text{and} \quad n_z^2 \equiv \frac{\varepsilon_z}{\varepsilon_0} = \frac{c^2}{u_z^2}, \quad \Rightarrow \quad \varepsilon(\omega) = \varepsilon_0 \begin{pmatrix} n_o^2(\omega) & 0 & 0 \\ 0 & n_o^2(\omega) & 0 \\ 0 & 0 & n_z^2(\omega) \end{pmatrix},$$

We call *positive uniaxial media* to those where $n_z > n_o$, while *negative uniaxial media* correspond to $n_z < n_o$.

For uniaxial media, Eq. (5.16) gets reduced to

$$(u_0^2 - u^2) [(s_x^2 + s_y^2)(u_z^2 - u^2) + s_z^2(u_0^2 - u^2)] = 0 \quad (5.19)$$

The objective then is to find the two possible solutions for the phase velocity u with which the electric field can propagate inside the anisotropic medium. Keeping in mind Eq.(5.17), this is equivalent to find the two possible refractive indexes that a wave can experience when propagating in this material:

1. Ordinary wave:

This solution corresponds to $(u_0^2 - u^2) = 0$. The resulting wave propagates with velocity u_o and refractive index n_o . Solving by components Eq. (5.13), we obtain the two conditions always fulfilled by the ordinary solution:

$$\vec{s} \cdot \vec{E}_o = 0 \quad E_{o,z} = 0 \quad \Rightarrow \quad \vec{S}_o \parallel \vec{s}$$

The ordinary component of the Poynting vector $\vec{S}_o$ is parallel to the direction of propagation, the corresponding electric field is perpendicular to it, and its refractive index is totally independent from it.

2. Extraordinary wave:

This solution corresponds to $[(s_x^2 + s_y^2)(u_z^2 - u^2) + s_z^2(u_0^2 - u^2)] = 0$. Defining

$$u_e^2 \equiv (s_x^2 + s_y^2)(u_z^2 - u^2) + s_z^2(u_0^2 - u^2) \quad (5.20)$$

it becomes now clear how the velocity of the extraordinary wave depends on the direction of propagation inside the medium. Again, from Eq.(5.17) this also implies that the *extraordinary refractive index* will depend on this direction, too:

$$n_e^2(\vec{s}) \equiv \frac{c^2}{u_e^2} = \frac{c^2}{(s_x^2 + s_y^2)u_z^2 + s_z^2u_0^2} = \frac{n_o^2n_z^2}{(s_x^2 + s_y^2)n_o^2 + s_z^2n_z^2} \quad (5.21)$$

or equivalently

$$n_e^2(\theta) = \frac{n_o n_z}{\sqrt{n_o^2 \sin^2 \theta + n_z^2 \cos^2 \theta}}, \quad (5.22)$$

where θ is the angle between the direction of propagation $\vec{s}$ and the optical axis. Varying this angle is what allows to match the velocity of the extraordinary wave to the one of the ordinary wave, accomplishing phase-matching and constructive interferences. The angle for which this occurs is the so-called *phase-matching angle* θ_m .

Solving again Eq.(5.13) by components, the conditions for the extraordinary wave are finally reached:

$$\vec{D}_e \cdot \vec{D}_o = 0 \quad \vec{s} \cdot \vec{D}_e = 0 \quad \Rightarrow \quad \vec{S}_e \nparallel \vec{s}$$

The anisotropic properties are now evident: the Poynting vector $\vec{S}_e$ is not parallel to the direction of propagation, which implies that energy and phase can propagate in different directions. Note how the electric field of the extraordinary component $\vec{E}_e$ is in general not parallel to the direction of propagation $\vec{s}$, since in this case $\vec{E}_e \nparallel \vec{D}_e$.

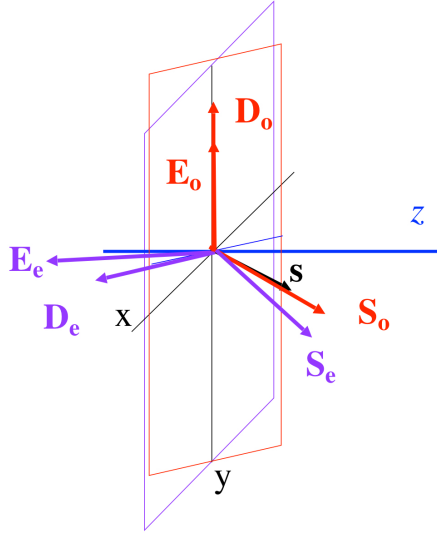


Figure 5.3: General scheme of an electromagnetic wave propagating in a uniaxial medium. Only the electric fields are shown for simplicity. Taken from [49].

Bibliography

- [1] Pierre-Marie Paul, Elena S Toma, Pierre Breger, Genevive Mullot, Frédérique Augé, Ph Balcou, Harm Geert Muller, and Pierre Agostini. Observation of a train of attosecond pulses from high harmonic generation. *Science*, 292(5522):1689–1692, 2001.
- [2] Albert Einstein. Über einen die erzeugung und verwandlung des lichtes betreffenden heuristischen gesichtspunkt, 1905.
- [3] Cristian Manzoni and Giulio Cerullo. Design criteria for ultrafast optical parametric amplifiers. *Journal of Optics*, 18(10):103501, 2016.
- [4] Prem B Bisht. *An Introduction to Photonics and Laser Physics with Applications*. IOP Publishing, 2022.
- [5] Robert W Boyd, Alexander L Gaeta, and Enno Giese. Nonlinear optics. In *Springer Handbook of Atomic, Molecular, and Optical Physics*, pages 1097–1110. Springer, 2008.
- [6] Valentin G Dmitriev, Gagik G Gurzadyan, and David N Nikogosyan. *Handbook of nonlinear optical crystals*, volume 64. Springer, 2013.
- [7] Günter Steinmeyer, Bastian Borchers, and Fabian Lücking. *Carrier-Envelope Phase Stabilization*, pages 89–110. Springer Berlin Heidelberg, Berlin, Heidelberg, 2013.
- [8] Zenghu Chang. *Fundamentals of attosecond optics*. CRC press, 2016.
- [9] R. Paschotta. Carrier-envelope offset. RP Photonics Encyclopedia. Available online at https://www.rp-photonics.com/carrier_envelope_offset.html, doi = 10.61835/ui4, urldate = 2024-05-19, keywords = carrier-envelope offset, CEO frequency, CEP, absolute phase.
- [10] Maximilian Bradler. *Bulk continuum generation: the ultimate tool for laser applications and spectroscopy*. PhD thesis, lmu, 2014.
- [11] T Imran and G Figueira. Efficient white-light continuum generation in transparent solid media using 250 fs, 1053 nm laser pulses. In *AIP Conference Proceedings*, volume 1228, pages 370–375. American Institute of Physics, 2010.
- [12] David Griffiths. *Introduction to elementary particles*. John Wiley & Sons, 2020.

- [13] Gihan Basnayake, Yasashri Ranathunga, Suk Kyoung Lee, and Wen Li. Three-dimensional (3d) velocity map imaging: from technique to application. *Journal of Physics B: Atomic, Molecular and Optical Physics*, 55(2):023001, 2022.
- [14] Mike Reid. *Velocity map imaging: from gases to surfaces*. The University of Manchester (United Kingdom), 2013.
- [15] David Staedter. *Femtosecond time-resolved spectroscopy in polyatomic systems investigated by velocity-map imaging and high-order harmonic generation*. PhD thesis, Université Paul Sabatier-Toulouse III, 2013.
- [16] André TJB Eppink and David H Parker. Velocity map imaging of ions and electrons using electrostatic lenses: Application in photoelectron and photofragment ion imaging of molecular oxygen. *Review of Scientific Instruments*, 68(9):3477–3484, 1997.
- [17] Dominik Schomas. *Helium and Neon nanoplasmas ignited by ultrashort NIR, MIR and x-ray laser pulses*. PhD thesis, Dissertation, Universität Freiburg, 2019, 2019.
- [18] Allan Maple de Oliveira. *Velocity-map Imaging Photoelectron Spectroscopy of Small Molecular Anions*. PhD thesis, University of Colorado at Boulder, 2018.
- [19] Daniel D Hickstein. *Photoelectron and photoion spectroscopy of atoms, nanoparticles, and nanoplasmas irradiated with strong femtosecond laser fields*. PhD thesis, University of Colorado at Boulder, 2014.
- [20] Dr. Andreas Wituschek. Velocity map imaging. *Fortgeschrittenen Praktikum*, 2016.
- [21] Dominik Schomas, Nicolas Rendler, Jakob Krull, Robert Richter, and Marcel Mudrich. A compact design for velocity-map imaging of energetic electrons and ions. *The Journal of chemical physics*, 147(1), 2017.
- [22] Christof Bartels. *Angular distributions of photoelectrons from cold, size-selected sodium cluster anions*. PhD thesis, Verlag nicht ermittelbar, 2008.
- [23] Vladimir Dribinski, Alexei Ossadtchi, Vladimir A Mandelshtam, and Hanna Reisler. Reconstruction of abel-transformable images: The gaussian basis-set expansion abel transform method. *Review of Scientific Instruments*, 73(7):2634–2642, 2002.
- [24] Gustavo A Garcia, Laurent Nahon, and Ivan Powis. Two-dimensional charged particle image inversion using a polar basis function expansion. *Review of Scientific Instruments*, 75(11):4989–4996, 2004.
- [25] Bernhard Dick. Inverting ion images without abel inversion: maximum entropy reconstruction of velocity maps. *Physical Chemistry Chemical Physics*, 16(2):570–580, 2014.
- [26] Marc JJ Vrakking. An iterative procedure for the inversion of two-dimensional ion/photoelectron imaging experiments. *Review of Scientific Instruments*, 72(11):4084–4089, 2001.

- [27] GM Roberts, JL Nixon, J Lecointre, E Wrede, and JRR Verlet. Toward real-time charged-particle image reconstruction using polar onion-peeling. *Review of Scientific Instruments*, 80(5), 2009.
- [28] Greif Michael Osterwalder Jürg Hengsberger Matthias Locher Reto Lucchini Matteo Gallmann Lukas Castiglioni, Luca and Ursula Keller. Novel insights into photoemission from solids: Surface rabbit yields absolute delays and reveals temporal structure beyond transport phenomena. Jul 2013.
- [29] Y Mairesse, A De Bohan, LJ Frasinski, H Merdji, LC Dinu, P Monchicourt, P Breger, M Kovacev, R Taïeb, B Carré, et al. Attosecond synchronization of high-harmonic soft x-rays. *Science*, 302(5650):1540–1543, 2003.
- [30] Praveen Kumar Maroju, Michele Di Fraia, Oksana Plekan, Matteo Bonanomi, Barbara Merzuk, David Busto, Ioannis Makos, Marvin Schmoll, Ronak Shah, Primož Rebernik Ribič, et al. Attosecond coherent control of electronic wave packets in two-colour photoionization using a novel timing tool for seeded free-electron laser. *Nature Photonics*, 17(2):200–207, 2023.
- [31] Joseph Duris, Siqi Li, Taran Driver, Elio G Champenois, James P MacArthur, Alberto A Lutman, Zhen Zhang, Philipp Rosenberger, Jeff W Aldrich, Ryan Coffee, et al. Tunable isolated attosecond x-ray pulses with gigawatt peak power from a free-electron laser. *Nature Photonics*, 14(1):30–36, 2020.
- [32] David Busto. *Quantum interference effects in attosecond photoionization dynamics*. Lund University, 2020.
- [33] FEMTOLASERS Produktions GmbH. *FEMTOPOWERTM V CEP4 USER MANUAL*.
- [34] Muhammad Jahanzeb. *Nonlinear temporal compression of femtosecond laser pulses using a stretched hollow-core fiber setup*. Master’s thesis. Albert-Ludwigs-Universität Freiburg, 2022.
- [35] R. Paschotta. Gain narrowing. RP Photonics Encyclopedia, May 2006. Available online at https://www.rp-photonics.com/gain_narrowing.html, url = <https://www.rp-photonics.com/gainnarrowing.html>, doi = 10.61835/uiB, urldate = 2024-04-06, keywords = gain narrowing, bandwidth, laser, amplifier, pulse duration, ultrashort pulses.
- [36] Sonal Saxena. *Studies on broadband terahertz radiation from ultra-short two-color laser induced plasma*. PhD thesis, 10 2019.
- [37] Hitachi Aberration-Corrected Concave Gratings for Flat-Field Spectrographs. *Hitachi Hightechnologies America, Inc.* https://www.hitachi-hightech.com/products/images/9797/ana-grating_05.pdf.
- [38] Lawrence Berkeley Laboratory. https://henke.lbl.gov/optical_constants/filter2.html.

- [39] Matteo Moiola. *Attosecond electron dynamics in noble gases and development of a high-repetition-rate OPCPA for coincidence spectroscopy*. PhD thesis, Dissertation, Universität Freiburg, 2023, 2023.
- [40] A. Kramida, Yu. Ralchenko, J. Reader, and NIST ASD Team. NIST Atomic Spectra Database (ver. 5.11), [Online]. Available: <https://physics.nist.gov/asd> [2024, June 1]. National Institute of Standards and Technology, Gaithersburg, MD., 2023,.
- [41] JJ Yeh. Atomic calculation of photoionization cross-section and asymmetry parameters gordon and breach science publishers. *Langhorne, PE (USA)*, 1993.
- [42] JJ Yeh and I Lindau. Atomic data and nuclear data tables, vol. 32, no. 1, 1985.
- [43] Rick Trebino. *Frequency-resolved optical gating: the measurement of ultrashort laser pulses*. Springer Science & Business Media, 2012.
- [44] R. Paschotta. Frequency-resolved optical gating. RP Photonics Encyclopedia. Available online at https://www.rp-photonics.com/frequency_resolved_optical_gating.html, doi = 10.61835/pda, urldate = 2024-05-18, keywords = frequency-resolved optical gating, FROG, pulse characterization.
- [45] R. Paschotta. Instantaneous frequency. RP Photonics Encyclopedia. Available online at https://www.rp-photonics.com/instantaneous_frequency.html, doi = 10.61835/ooz, urldate = 2024-06-01, keywords = instantaneous frequency, time-dependent frequency, chirp, PLL, VCO.
- [46] Eric Moon, He Wang, Steve Gilbertson, Hiroki Mashiko, Michael Chini, and Zenghu Chang. Advances in carrier-envelope phase stabilization of grating-based chirped-pulse amplifiers. *Laser & Photonics Reviews*, 4(1):160–177, 2010.
- [47] Xiaoming Ren, AM Summers, Aram Vajdi, Varun Makhija, CW Fehrenbach, Nora G Kling, KJ Betsch, Z Wang, MF Kling, KD Carnes, et al. Single-shot carrier-envelope-phase tagging using an f–2f interferometer and a phase meter: a comparison. *Journal of Optics*, 19(12):124017, 2017.
- [48] Anne Harth, Nicolas Douguet, Klaus Bartschat, Robert Moshhammer, and Thomas Pfeifer. Extracting phase information on continuum-continuum couplings. *Physical Review A*, 99(2):023410, 2019.
- [49] Prof. Dr. Carlos Hernández García. Óptica II Lecture. *Universidad de Salamanca*, 2023.
- [50] Louis De Broglie. The wave nature of the electron. *Nobel lecture*, 12:244–256, 1929.

