

Attosecond light pulses for the study of spin dynamics

Master thesis

presented by

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Summary

This internship focuses on the generation of high harmonics for spin- and angle-resolved photoemission spectroscopy (ARPES), addressing challenges such as long acquisition times and high repetition rates. A Ytterbium (Yb)-based femtosecond laser with a central wavelength of 1030 nm was utilized, and post-compression techniques were employed to enhance its performance.

The project also involved exploring the theory behind light-carrying orbital angular momentum and conducting experiments to investigate Spin-Orbit coupling in non-linear processes. Preliminary results showed variations in the spectrum during self-phase modulation, suggesting possible evidence of this coupling, though further experiments are needed for confirmation.

The thesis is organized as follows: Section 2 covers the theory behind post-compression and its application using a Multi-Pass Cell. It also explains the principle of high harmonic generation through the three-step model. Section 3 details the experimental implementation of post-compression, where the pulse was spectrally broadened and thus its duration was decreased. We used Frequency-Resolved Optical Gating (FROG) to measure the spectral phase and pulse duration of the laser beam. The Harmonics were generated in optimal conditions. This involved taking a series of measurements under varying energy and pressure.

Section 4 explores the principles of light carrying orbital angular momentum, explained in the paraxial optics regime. It describes how this light beam was generated and includes details about experiments using this light, such as beam profiling, nonlinear compression of the beam, and polarization analysis. We also conducted experiments to see the evidence of orbital angular momentum. These experiments aimed to observe variations in the broadening of the beam after undergoing nonlinear effects.

These efforts represent progress in post-compression techniques and the generation of high harmonics. This work sets the stage for future advancements in ultrafast spectroscopy and high-resolution imaging, deepening our understanding of electron dynamics.

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1 Introduction

The observation of events in the time domain requires very different techniques depending on their time scales. For example, a heartbeat lasts around one second, while nuclear dynamics in molecules occur on the picosecond to attosecond scale ($10^{-12} - 10^{-18}$ s), the latter being the natural time scale of electron dynamics. The development of ultrafast light sources in the last two decades has revolutionized our ability to study these rapid processes. This involves probing into electron dynamics that are difficult to observe experimentally and as well as to be described theoretically. Attosecond sources, based on High Harmonic Generation (HHG), have opened new frontiers in dynamic XUV spectroscopy on the sub-femtosecond scale, as compared to Free Electron Lasers (FELs) and synchrotrons. They both play a pivotal role in advancing our understanding of ultrafast science, but both technologies have their limitations. They are large-scale facilities, which are expensive to build and operate, while HHG source is a tabletop system, which is more accessible and offers a superior temporal resolution, which is crucial for observing electron dynamics with high precision.

The discovery of the High Harmonic Generation in the late 1980s was pivotal for entering the attosecond regime. This discovery was made during experiments with early high-intensity laser sources ($10^{13} - 10^{15}$ W/cm²) [1, 2]. HHG involves ionizing atoms with a laser field, accelerating free electrons, and recombining them with their parent ions to emit high-energy photons, producing coherent XUV radiation essential for high-resolution spectroscopy and imaging.

The development of attosecond tools is continuously leading to the introduction of new spectroscopic and measurement methods, which will offer the opportunity to investigate unexplored research areas with high time resolution. Attosecond sources are now employed in various applications, for instance in absorption and reflectivity measurement, [3], in circular dichroism measurements [4], time- and angle-resolved photoemission spectroscopy (trARPES) [5] and element-specific magneto-optic Kerr effect (EUV-MOKE) spectroscopy. [6].

Historically, Titanium Sapphire (Ti:Sa) lasers have dominated sub-50-fs pulse duration and high-intensity applications. Despite their limitations in repetition rates, they have provided sufficient intensity for many experiments. However, their limited average power and repetition rates restrict their applicability in experiments requiring long acquisition times. Thus recently, Ytterbium (Yb) ion-doped lasers have been increasingly used due to better specifications which involve a high repetition rate, and ease of management. However, their narrower emission bandwidth necessitates advanced pulse compression techniques to achieve the desired pulse durations, and high power management which must be addressed in experimental setups.

The objective of this internship is to generate and optimize a High Harmonic Generation (HHG) source tailored for spin-resolved ARPES, which requires long acquisition times and high repetition rates. We utilized a new Yb-based femtosecond laser source of 330 fs, which was post-compressed in a multi-pass cell (MPC). The compressed pulse was used for the generation of high harmonics.

Finally, this new laser source was used to generate a light carrying angular momentum. This was achieved using polarization-shaping or wavefront-shaping tailored optics. We investigated the effect of light carrying Orbital and Spin Angular momentum in a Multi-Pass Cell.

2 Theoretical Framework for the Post Compression and High Harmonic Generation

2.1 Post compression

To generate attosecond pulses through high-order harmonic generation (HHG), laser pulses need to be extremely short and intense. Typically, HHG requires peak intensities around 10^{14} W/cm^2 at focus, which demands very short pulse durations. Thus, an ideal regime for HHG is to use pulses of 20-50 fs duration. However, many laser sources, such as those based on Ytterbium (Yb), inherently produce longer pulses, often spanning several hundred femtoseconds.

To address this, post-compression techniques are employed. One widely used method involves self-phase modulation, a non-linear optical effect that, under suitable conditions, broadens the pulse's spectrum as it travels through a non-linear medium. This process is critical for compressing longer pulses to the desired duration. To understand the processes involved, we introduce some fundamental theories in non-linear optics.

2.1.1 Electric polarization and Wave equation

The propagation of electromagnetic waves in a free space, that is no free charge and nonmagnetic medium, can be represented using Maxwell's equation:

$$\nabla \cdot \mathbf{D} = 0 \quad (1)$$

$$\nabla \cdot \mathbf{B} = 0 \quad (2)$$

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \quad (3)$$

$$\nabla \times \mathbf{H} = \frac{\partial \mathbf{D}}{\partial t} \quad (4)$$

where $\mathbf{E}$ and $\mathbf{B}$ are the electric and the magnetic field respectively, $\mathbf{H}$ is the magnetic field intensity, $\mathbf{D}$ is the electric displacement vector, and is given by the formula:

$$\mathbf{D} = \epsilon \mathbf{E} + \mathbf{P} \quad (5)$$

where $\mathbf{P}$ is the dielectric polarization density, and $\epsilon = \epsilon_0 \epsilon_r$ is the absolute permittivity with ϵ_0 being the permittivity of free space and ϵ_r the relative permittivity.

Solving the Maxwell equation for an isotropic, dispersionless medium, we get the wave equation for a monochromatic field,

$$\nabla^2 \mathbf{E} = \mu_0 \frac{\partial^2 \mathbf{D}}{\partial t^2} \quad (6)$$

To describe optical linearity, we need to study how the polarization $\mathbf{P}$ of a material depends on the strength of the electric field applied. In linear optics, the induced polarization depends linearly on the electric field strength, described by the relation:

$$\mathbf{P}^{(1)}(t) = \epsilon_0 \chi^{(1)} \mathbf{E}(t) \quad (7)$$

where the constant of proportionality $\chi^{(1)}$, known as linear susceptibility, is a second-rank tensor. In nonlinear optics, this polarization can be expressed as a power series in the field strength $\mathbf{E}(t)$ as:

$$\mathbf{P}(t) = \epsilon_0 [\chi^{(1)} \mathbf{E}(t) + \chi^{(2)} \mathbf{E}^2(t) + \chi^{(3)} \mathbf{E}^3(t)].. \quad (8)$$

$$= \mathbf{P}^{(1)}(t) + \mathbf{P}^{(2)}(t) + \mathbf{P}^{(3)}(t) \dots \quad (9)$$

where $\chi^{(2)}$ and $\chi^{(3)}$ are second and third order nonlinear optical susceptibilities respectively. We can write the polarization as

$$\mathbf{P}(t) = \mathbf{P}^{(1)}(t) + \mathbf{P}^{(NL)} \quad (10)$$

This can be further substituted in the equation for displacement vector and it becomes:

$$\mathbf{D} = \epsilon \mathbf{E}(t) + \mathbf{P}^{(1)}(t) + \mathbf{P}^{(NL)} \quad (11)$$

Thus we can rewrite the solution of the Maxwell equation (6)[7] as:

$$\nabla^2 \mathbf{E} - \frac{\epsilon_r \mu_r}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \mathbf{P}^{NL}}{\partial t^2} \quad (12)$$

The post-compression process and HHG are most efficient in gases, particularly rare gases like Argon, which is a centrosymmetric medium. While high harmonics can also be generated in non-centrosymmetric gas molecules like N_2 , rare gases are preferred due to their higher efficiency.

The non-linear effects that we study can change depending on the medium symmetry. It should be noted that even order non-linear optical interactions occur in noncentrosymmetric media- those are the mediums that do not display inversion symmetry (See Fig- 1)

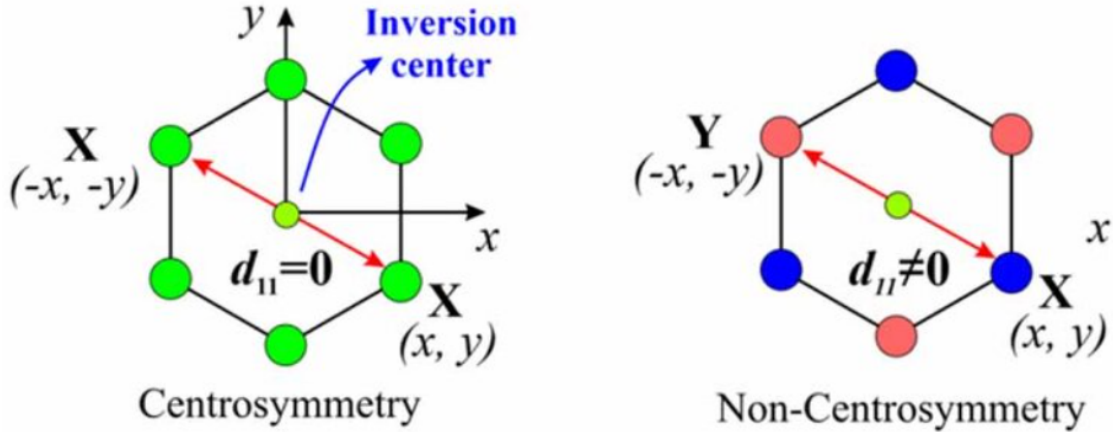


Figure 1: Comparison between centrosymmetric and non-centrosymmetric crystal structure in 2D materials (a) There is an inversion center at the point between (x, y) and $(-x, -y)$ (b) No inversion center . [8]

In liquids, gases, and amorphous solids that display inversion symmetry, $\chi^{(2)}$ vanishes. This case is demonstrated for second harmonic generation in a medium that responds instantaneously to the applied electric field. Consider the nonlinear polarization given by:

$$\mathbf{P}(t) = \epsilon_0 \chi^{(2)} \mathbf{E}^2(t) \quad (13)$$

where the applied electric field is $\mathbf{E}(t) = \mathbf{E}_0 \cos \omega t$. Now, changing the sign of the electric field $\mathbf{E}(t)$, leads to the change in the sign of the induced polarization since our assumption was the medium has inversion symmetry. Hence the relationship becomes

$$-\mathbf{P}(t) = \epsilon_0 \chi^{(2)} [-\mathbf{E}(t)]^2 \quad (14)$$

$$-\mathbf{P}(t) = \epsilon_0 \chi^{(2)} [\mathbf{E}(t)]^2 \quad (15)$$

We see that $\mathbf{P}(t)$ must be equal to $-\mathbf{P}(t)$ which can occur only if $\mathbf{P}(t)$ vanishes identically. And this shows

that:

$$\chi^{(2)} = 0 \quad (16)$$

This argument can be extrapolated to all even-order nonlinear susceptibility. However, third-order nonlinear optical interaction can occur in both centrosymmetric and noncentrosymmetric media, which is what we will be using as our medium for the experiments conducted.

2.1.2 Self-Phase modulation

As mentioned before, when a short optical pulse passes through a dispersive medium, some nonlinear optical effects can occur. The nonlinear process of self-phase modulation modifies the spectral content of the pulse. In general, both self-phase modulation and group velocity dispersion (linear effect) occur simultaneously, and modify the shape of the optical pulse.

Self-phase modulation (SPM) occurs when a pulse traveling through a medium induces a refractive index change in that medium, leading to a phase modulation of the pulse itself and this leads to a change in the frequency spectrum of the pulse. Self-phase modulation was first proposed to explain a spectrally broadened output from the self-focusing of a Q-switched laser pulse in a liquid with a large optical Kerr constant. In this case, the spectral broadening was on the order of 0.0124 eV. [9].

The principle lies in the fact that a short laser beam can induce an intensity-dependent refractive index change in the medium. This is a nonlinear change in the refractive index. In turn, the modified refractive index imposes a time-dependent phase change onto the driving laser, hence the name "self-phase modulation". As we will see, this leads to a spectral broadening in the time domain.

To understand the origin of this effect, we consider the propagation of an optical pulse with frequency ω in a medium:

$$\mathbf{E}(z, t) = \mathbf{A}(z, t)e^{i(kz - \omega t)} + c.c \quad (17)$$

where $k = n\omega/c$ is the dispersion relation in a linear medium. We consider the third-order nonlinear term driven by this pulse and is given by [10]:

$$\mathbf{P}^{(3)}(r, t) = 3\epsilon_0\chi^3(\omega : \omega, -\omega, \omega)|\mathbf{A}|^2\mathbf{A}e^{i(kz - \omega t)} \quad (18)$$

where $\chi^3(\omega : \omega, -\omega, \omega)$ is the Kerr susceptibility. Here this equation automatically satisfies the phase-matching condition. Indeed, the non-linear polarization wave vector can be written as $k - k + k = k$ which is the same as the wave vector of the incident electromagnetic field.

Here, $\mathbf{A}(z, t)$ is the pulse envelope which we assume varies slowly in time and space at the scale of the optical period T_0 and the wavelength λ_0 [11]:

$$\mathbf{A}(z, t + T_0) \approx \mathbf{A}(z, t) \quad (19)$$

$$\mathbf{A}(z + \lambda_0, t) \approx \mathbf{A}(z, t) \quad (20)$$

so that we can apply the condition

$$\frac{\partial^2 A}{\partial z^2} \ll k \frac{\partial A}{\partial z} \quad (21)$$

This assumption is called the slowly varying envelope approximation (SVEA). Applying this to equation (12), we get the non-linear propagation equation[11]:

$$\frac{\partial \mathbf{A}}{\partial z} = i \frac{\omega}{2\epsilon_0 n c} \mathbf{P}^{(3)}(r, t) \quad (22)$$

We now consider linearly polarized light, which allows us to project all vectorial quantities onto the

polarization axis of the pulse in order to obtain a scalar equation [10]:

$$P(\mathbf{r}, t) = \epsilon_0 \left[\chi^{(1)} + 3\chi^{(3)} |A(\mathbf{r}, t)|^2 \right] A(\mathbf{r}, t) e^{i(kz - \omega t)} \quad (23)$$

$$\equiv \epsilon_0 \chi^{(eff)} A(\mathbf{r}, t) e^{i(kz - \omega t)} \quad (24)$$

Here the Kerr effect can be written as an effective susceptibility $\chi^{(eff)}$, which has a dependence on the intensity of light. We can therefore define an intensity-dependent refractive index [10]:

$$n(I) = (1 + \chi^{(eff)}(I))^{(1/2)} \quad (25)$$

where we have the linear refractive index has:

$$n_0 = (1 + \chi^{(1)}(I))^{(1/2)} \quad (26)$$

thus introducing the nonlinear refractive index as:

$$n(t) = n_0 + n_2 I(t) \quad (27)$$

where n_2 is the second-order refractive index and defined by:

$$n_2 = \frac{3\chi^{(3)}(\omega, -\omega, \omega)}{4\epsilon_0 n_0^2 c} \quad (28)$$

The nonlinear refractive index has its dimension as the inverse of the intensity and is commonly given in units of cm^2/W . The effect of the refractive index changing with light is often called the Kerr effect.

Due to the change in the refractive index, as seen in equation (27), the change of phase of the transmitted pulse is given by [7]:

$$\phi_{NL}(t) = -n_2 I(t) \omega_0 \Delta z / c \quad (29)$$

where Δz is the distance that pulse has propagated, and ω_0 is the central frequency. Now the total phase accumulated can be written as:

$$\phi(t) = \omega_0 t - \frac{\omega_0}{c} n_0 \Delta z + \phi_{NL}(t) \quad (30)$$

The instantaneous frequency considering this temporal phase is given by:

$$\omega(t) = \frac{d\phi(t)}{dt} = \omega_0 + \delta\omega(t) \quad (31)$$

$$\omega(t) = \omega_0 \left\{ 1 - \frac{\Delta z n_2}{c} \frac{dI(t)}{dt} \right\} \quad (32)$$

where $\delta\omega(t)$ is given by equation (33):

$$\delta\omega(t) = \frac{d}{dt} \phi^{NL}(t) \quad (33)$$

We will now consider the case of an ultrashort Gaussian pulse. The intensity at a time t can be expressed as:

$$I(t) = I_0 e^{-\frac{t^2}{\tau^2}} \quad (34)$$

where I_0 and τ are the peak intensity and half-width at $1/e$. Now, we can substitute the derivative of the

equation. 34 into equation. 32, and we have

$$\omega(t) = \omega_0 + \frac{2\Delta z n_2}{c\tau^2} t I_0 e^{-\frac{t^2}{\tau^2}} \quad (35)$$

Fig. 2 shows $\omega(t)$, displaying the frequency shift of each part of the pulse. It can be observed that the front part of the curve shifts towards low frequency, and the back part of the pulse to higher frequencies, while the peak of the pulse remains unchanged. In the center of the pulse, which is between, $t = \pm\tau/2$, we see a linear frequency shift, also called the chirp.

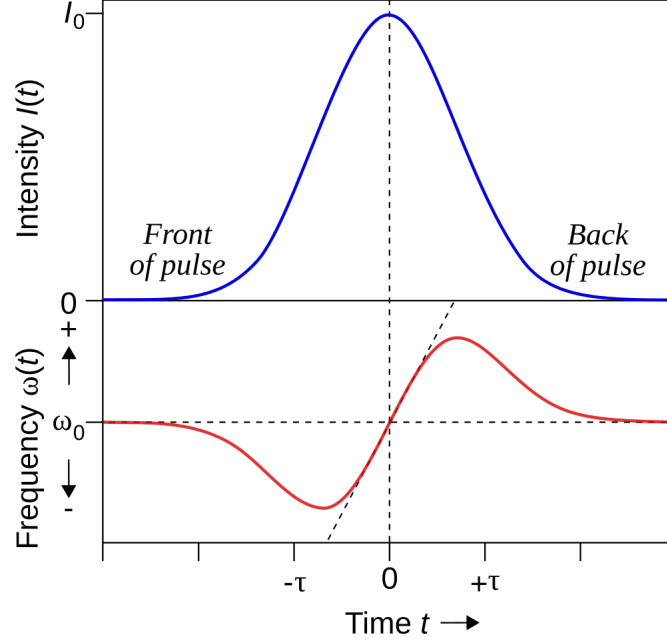


Figure 2: Instantaneous frequency shift in self-phase modulation. The top panel shows the Gaussian intensity distribution of the pulse. The bottom panel represents the frequency shift of the pulse resulting from self-phase modulation. In this bottom figure, the dashed line at the center represents the chirp.[12]

Consider ϕ_{NL}^{max} to be the maximum nonlinear phase shift obtained during propagation in the nonlinear medium. The spectrum will thus be broadened to a width of $\Delta\omega = \frac{\phi_{NL}^{max}}{T}$, where T is the pulse duration.[7] Fig. 3 represents an example of the modification of a laser spectrum by SPM.

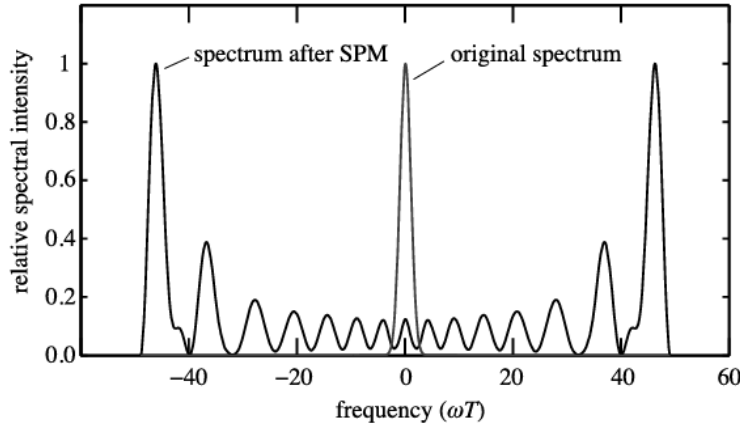


Figure 3: Example of broadening by SPM, considering a Gaussian input pulse, with a peak intensity producing $\phi_{NL}^{max} = 30\pi$ radian of nonlinear shift. T is the pulse half width to the e^{-2} intensity point.

The spectral width after broadening is equal to $30\pi \simeq 100$ in the dimensionless unit. [7]

We introduce a new quantity known as the B-integral, which represents the accumulated nonlinear

phase. The B-integral can be expressed in integral form as[13]:

$$B = \frac{2\pi}{\lambda} \int n_2 I_p(z) dz \quad (36)$$

where $I_p(z)$ denotes the peak intensity along the z-axis, λ is the central wavelength. Another equation of B-integral which is $B = \frac{2\pi}{\lambda} \int n_2 P / A_{eff} dz$, with A_{eff} which is the effective mode area, and P represents the peak power.

For a Gaussian beam, the B-integral simplifies to: [13]

$$B = 2\phi_{max}^{NL} \quad (37)$$

Another important quantity is the temporal compression ratio K which is related to the spectral broadening. Spectral broadening is inversely related to temporal broadening and K is defined as the ratio of the full width at half-maximum (FWHM) duration of the input pulse to that of the compressed pulse. For an initially Gaussian pulse and a resulting SPM broadened Fourier transform limited pulse, the compression ratio can be expressed as[13]:

$$K = \frac{B}{\pi} \quad (38)$$

which shows the direct proportionality between the B-integral and the temporal compression ratio, where an increase in the accumulated nonlinear phase (B-integral) leads to a higher degree of temporal compression.

2.1.3 Multi-pass cell for post-compression

To implement post compression, we use a multi-pass cell (MPC) where self-phase modulation (SPM) occurs several times in a nonlinear medium such as a bulk material or a gas placed at the center of the cell and use a negative dispersion optical setup in the subsequent propagation. By employing the cavity geometry, we can achieve higher broadening by repeating the process several times.

A gas-filled cavity is more advantageous compared to a solid-state medium because, during propagation in a bulk medium, there would be optical interface losses, the spatial Kerr effect lead to beam degradation and spatiotemporal coupling, and these would reduce the accumulation of a large temporal nonlinear phase [14]. In this section, we introduce the basic concept of MPC and describe the processes occurring inside a Herriott-type MPC cell as the beam propagates through it.

The setup consists of two identical mirrors facing each other with a radius of curvature R , The mirrors are separated by a distance L . The Herriott configuration allows for a constant beam profile to be maintained over several round trips in the multi-pass cell (MPC), and we get stationary beams in the cell.

The maximum distance between the mirror is twice the radius of the curvature of the mirror. There are several configurations as the cavity length L increases while maintaining a fixed radius of curvature R

- When the cavity length L is less than twice the Rayleigh distance $2z_R$, which means it is small as compared to R , the beam radius (ω_0) is also small. The Rayleigh distance z_R for a Gaussian beam is given by:

$$z_R = \frac{\pi \omega_0^2}{\lambda} \quad (39)$$

where λ is the wavelength of the medium. It is the distance from the beam waist along the propagation axis to the point where the beam radius has increased by a factor of $\sqrt{2}$

- In confocal cavity $L = R$, the beam has the largest waist radius. The propagation distance in this configuration is twice the Rayleigh distance.
- For values of L approaching cavity stability, $L = 2R$, the waist radius tends to zero, while the beam size on the mirrors increases towards infinity

The relationship between the beam waist and the beam size on the mirror is given by [15]:

$$\left(\frac{w_m}{w_o}\right)^2 = \frac{2R}{2R - L} \quad (40)$$

We work in the regime where $L \leq 2R$, which is when the waist w_0 is minimum with maximum beam size on the mirrors. The MPC is similar to a laser cavity (used to amplify the light by multiple reflections). The Herriott cell forms transverse modes that can propagate within the cavity without changing shape, except for scaling or phase shifts. This setup preserves the Gaussian beam q-parameter of the input beam which is mode-matched to the MPC eigenmode, where the q-parameter is a complex number describing the properties of a Gaussian beam at a particular point along the propagation direction. As it is mode-matched, it leads to a constant B-integral at each round trip.

A typical MPC setup used for post-compression is depicted in Fig - 4. Before the cell, the input laser beam is mode-matched to the given mode through a telescope and ensures the beam property remains the same in each pass of the cell. Further, the beam is coupled to the MPC off-axis via a small injection mirror for the input beam, and after a targeted number of round trips the spectrally broadened output beam is coupled out through an extraction mirror. The spectrally broadened beam is extracted for after a chosen number of passes (the 27th in our case). After the extraction, the beam needs to be collimated, which is done through a telescope (will be described in Section 3), and then passes through a chirped mirror compressor to remove SPM-induced chirp by providing negative dispersion.

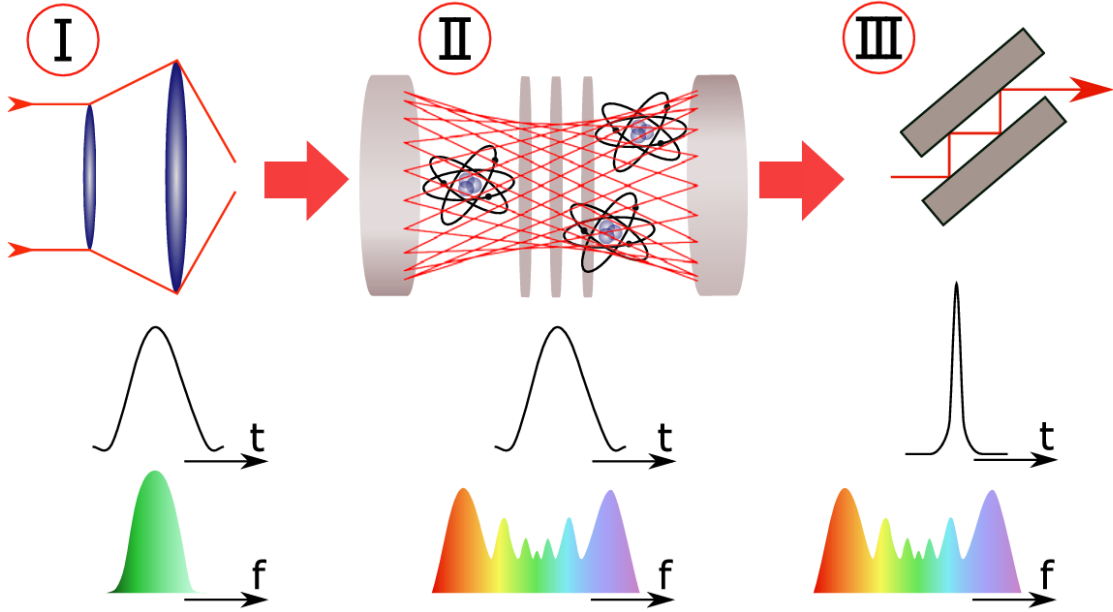


Figure 4: Post compression in MPC: Step I is the mode matching, Step II: Spectral broadening in MPC, Step III: Temporal compression using a chirped mirror compressor. [13]

Simulation conducted in the lab for $800\mu J$ energy, with argon for pressure of 3bar has a total B-integral that reached 11.76π and a corresponding pulse duration of $\tau_{out} = 29.76$ fs at the output of MPC. An illustration of how the multiple reflections will be seen in the MPC is shown in Fig.5. In the experimental implementation presented further, we will use 27 passes.

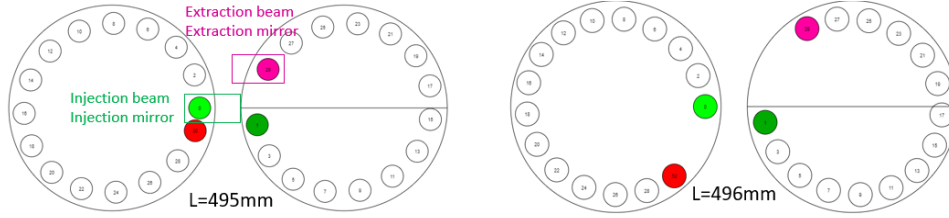


Figure 5: Beam pattern at the MPC mirror, for two different lengths of the cavity. The configuration in the lab (left), where the beam is extracted for 29th pass, and when the length increases (right), the number of possible passes increases with a smaller focus.[16]

2.2 High Harmonic Generation

The strong field regime is defined as the regime when the electric field component of the light becomes comparable to the coulomb field of atoms or molecules. Modern ultrafast laser technology provides access to this regime. One of the main phenomena that was discovered as a result of this access is high harmonic generation.

Interaction of intense laser pulses with a target medium, results in the emission of photons with frequencies that are integer multiples (odd harmonics) of the driving laser frequency. It is a highly nonlinear process, and the spectrum generated extends over several tens to hundreds of eV, thus making it the perfect candidate for generating spectra ranging from XUV to soft X-ray radiation. With an appropriate phase relationship, an HHG spectrum of discrete harmonics corresponds to the attosecond pulse train (APT) in the domain.

In this chapter, we will present the basic theory behind HHG. This constitutes describing the HHG from a semi-classical point of view, which is also called the three-step.

2.2.1 Semiclassical description: Three-step model

The three-step model, introduced in 1993, [17, 18] can be used to describe how high harmonics are generated during an interaction between a single atom of a noble gas and a laser pulse.

- Ionization: The laser field detaches an electron from the valence shell by tunnel ionization
- Excursion in the continuum: Free electrons are accelerated by the laser field
- Radiative recombination: The kinetic energy gained by the electron is released through the emission of photons, during the recombination with the parent ion.

In the following section, we will elaborate on these steps:

2.2.1.1 Step 1: Tunneling Ionization An important quantity to introduce is the ponderomotive energy which is defined as the total oscillating energy acquired by an electron in an oscillating electromagnetic field, such as a laser field [19] .

$$U_p = \frac{e^2 E_0^2}{4m\omega^2} = \frac{e^2 I}{2c\epsilon_0 m\omega^2} \quad (41)$$

where e and m is the charge and the mass of the electron, E_0 is the electric field amplitude, ω is the angular frequency of the laser field. In the second part of the equation, I is the laser intensity, ϵ_0 is the dielectric constant in vacuum, and c is the speed of the light. Rewriting this equation in units more adequate for strong-field laser physics, we obtain:

$$U_p[eV] = 9.22 \times 10^{-20} \times (\lambda[nm])^2 \times I[W/cm^2] \quad (42)$$

It can be seen that ponderomotive energy has a quadratic and linear dependence on wavelength and laser intensity, respectively.

We consider a field with frequency such that $\hbar\omega_0 < I_p$, where I_p is the ionization energy of the atom, making single-photon ionization impossible. Two different regimes characterize the ionization of an atom in the optical field. This is the common case with a visible laser and a neutral atom.

At higher intensities, even though a single photon does not have enough energy to ionize the atom, the atom can absorb multiple photons simultaneously. The combined energy of these multiple photons can reach or exceed the ionization energy I_p , allowing the electron to be freed from the atom. This process is called multiphoton ionization (MPI). In MPI, the electric field of the laser is strong enough to facilitate the absorption of several photons, but it is not yet strong enough to distort the atomic potential significantly.

When the intensity of the optical field continues to increase, the electric field strength of the laser becomes comparable to the atomic Coulomb potential. In this regime, the laser field distorts the atomic potential barrier to such an extent that the electron can tunnel through this barrier and escape from the atom. This process is referred to as tunnel ionization.

The dependence of the ionization probability on the ionization potential and laser radiation, that is the intensity I and frequency ω was theoretically investigated by L. V. Keldysh. He introduced the Keldysh parameter, γ which defines the transition between the two ionization regime:

$$\gamma = \sqrt{\frac{I_p}{2U_p}} \quad (43)$$

Suppose the ponderomotive energy is greater than ionization energy. In that case, the potential well interacts with a strong linearly polarized electric field from a laser that is, $\gamma < 1$, the net potential is tilted, allowing the electron to tunnel into the continuum. Thus the tunneling regime dominates. Multiphoton ionization (MPI) dominates when $\gamma > 1$, which implies the ionization potential is greater than the ponderomotive energy. If the number of photons multiplied by the energy of each photon is greater than the ionization potential then the electron is freed. In a low-intensity field, electrons enter the continuum with the minimum number of photons required to free the electron. [20]

Here we will focus on the tunneling ionization regime, where we can have high harmonic generation. Consider an isolated atom in its ground state, with the Coulombic potential subjected to the ionic core given by $V_0(x) = -\frac{1}{|x|}$ Fig. 6 where x is the distance between the electron and the nucleus.

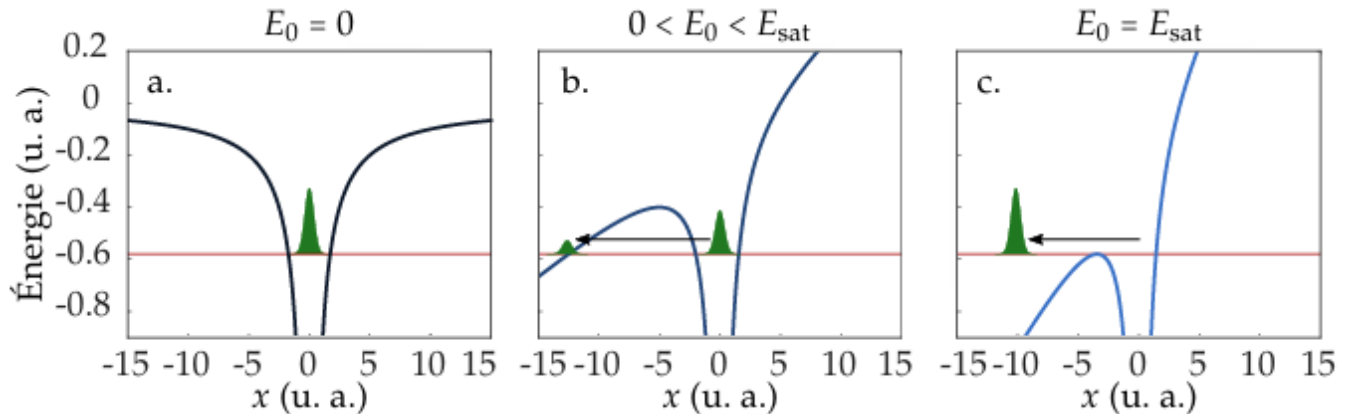


Figure 6: Potentials felt by the electron in the case of argon. $I_p = 15.76 \text{ eV} = 0.58 \text{ a.u.}$ (represented by the dotted line), (a) in the absence of an electric field (b), in the presence of a field $E_0 = 0.04 \text{ a.u.}$, where $\gamma < 1$ (c), and in the presence of a field at the over-the-barrier intensity of argon $E_0 = E_{sat} = 0.084 \text{ a.u.}$

Figure taken from [21].

For an electron in the ground state with an energy I_p below the ionization level, we introduce a monochromatic linearly polarized electric field $E(t) = E_0 \cos(\omega_0 t) \hat{x}$, where E_0 is the field amplitude and the angular frequency ω . The associated potential due to the electric field will be $E_0 x$ at the time of

origin. Thus, the total potential felt by the electron is given by :

$$V(x) = -\frac{1}{|x|} + E_0 x \quad (44)$$

As the laser field increases, the potential barrier lowers, allowing a portion of the electron wave packet to tunnel through the barrier (Fig -6b). The tunneling rate depends on the height and width of the potential barrier. The barrier-suppressed ionization regime is reached when the laser field is sufficiently high. In this regime, the electron can escape the potential directly, leading to the atom's ionization. We note this energy as E_{sat} , with corresponding saturation intensity I_{sat} . To obtain the I_{sat} , we take the derivative of Eqn: (44) and we look for coordinates $(x_m, V(x_m))$, where x_m is the distance at which the potential is maximum:

$$\frac{\partial V}{\partial x} = \frac{1}{x^2} + E_0 \quad (45)$$

and solving for x, we get "

$$x_m = -1/\sqrt{E_0} \quad (46)$$

with the corresponding potential at this position:

$$V(x_m) = -2\sqrt{E_0} \quad (47)$$

Since E_{sat} is the value at which the potential barrier is low enough for the electron to escape, and in this instance, the height of the barrier is equal to the ionization potential, we get,

$$V(x_m) = -I_p \implies I_{sat} = \frac{I_p^4}{16} \quad (48)$$

This intensity can be expressed in conventional units as $I_{sat} = 4 \times 10^{10} I_p^4 [eV]$ For tunnel ionization to occur, the intensity used must be lower than the saturation intensity, typically on the order of 10^{14} . The duration for which the potential barrier will be lowered is also important. This tunneling duration τ is characterized by the Keldysh parameter.

2.2.1.2 Step 2: Excursion in the continuum The dynamics of the electron in the strong laser field can be described by considering that the influence of the atomic Coulomb potential on the freed electron is negligible. In this case, the electron propagates in the continuum, governed by Newton's classical equation of motion for a charged particle in the electric field. This equation of motion is given by:

$$m\ddot{x} = -eE(t) = -eE_0 \cos(\omega t) \quad (49)$$

Here, $E(t)$ is the electric field of the laser, which accelerates the electron, allowing it to acquire kinetic energy. For the initial condition, we assume that $x(t_i) = 0$ and $\dot{x}(t_i) = 0$, where t_i is the time of ionization. This implies that we neglect the movement of the electron through the barrier, that is, it enters the continuum at the origin, and we assume the electron's velocity is zero as it crosses the barrier. By integrating this equation, we can follow the trajectory of the free electron in the continuum. The velocity $v(t)$ and the position $x(t)$ of the electron are given by:

$$\dot{x}(t) = v(t) = -\frac{eE_0}{m\omega} [\sin(\omega t) - \sin(\omega t_i)] \quad (50)$$

$$x(t) = \frac{eE_0}{m\omega^2} [\cos(\omega t) - \cos(\omega t_i)] + \frac{eE_0}{m\omega} \sin(\omega t_i)(t - t_i) \quad (51)$$

The first term represents the oscillatory motion of the electron. and the second term is a line, corresponding to the constant drift velocity, which grows linearly with time. After the laser pulse has passed,

the oscillatory term averages out to zero and only the drift motion remains.

2.2.1.3 Step 3: Radiative recombination After the electron is ionized and accelerated by the laser field in the continuum, it may cross the origin again and return to the ionic core. This trajectory leads to high harmonic emission. The direction of propagation itself does not play a role in the recombination of the electron, it happens when the electron is moving in the direction of the oscillating electric field. When the electron returns to the nucleus with non-zero kinetic energy, it drops back to the ground state, releasing energy. This process is called radiative recombination, as the electron gives back its energy.

To calculate the times at which the electron returns to the ionic core, we introduce the recombination time t_r , defined such that $x(t_r) = 0$. If $E(t_r)$ is the energy acquired by the freed electron in the continuum, then according to the conservation of energy, the energy of the photon emitted during the recombination event is given by [22]:

$$\hbar\omega = I_p + E(t_r) \quad (52)$$

where ω is the pulsation of the emitted photon, and $E(t_r)$ is given by:

$$E(t_r) = 2U_p[\sin(\omega t_r) - \sin(\omega t_i)]^2 \quad (53)$$

For different ionization times t_i , the electron can follow a trajectory that crosses the $x = 0$ position more than once. However, in practice, the photoelectron spreads out in the transverse direction as it propagates, so we neglect the contribution of multiple recollision events. The slope of the trajectory at the point where it crosses $x = 0$ determines the kinetic energy at the instant of the recombination. For each ionization time, there is a corresponding energy at a specific recombination time. The plot for $E(t_r)$ is shown in Fig-7 (b)

The maximum kinetic energy that an electron can acquire in the continuum is given by $E(t_r) = 3.17U_p$, which can be further used to predict the maximum photon energy, known as the cut-off energy, which is expressed as:

$$E_{cutoff} = I_p + 3.17U_p \quad (54)$$

In the context of high-harmonic generation, electrons can follow two distinct trajectories, referred to as the long and short trajectories, both resulting in the same energy but with different recombination times. This can be seen in Fig- 7 (a). The short trajectory corresponds to a propagation time that increases with harmonic order, while the long trajectory corresponds to a propagation time that decreases with harmonic order. The two trajectories converge and become indistinguishable at the cut-off energy. Fig- 8 gives an illustration of the three-step model in HHG.

The three-step process is repeated periodically every half cycle of the laser field, thus it has a periodicity of $T/2$ where $T = 2\pi/\omega_0$, which is the period of the fundamental laser. As a result of this temporal periodicity, the process has a $2\omega_0$ periodicity in the frequency domain. For a long enough generation pulse (several optical cycles), the spectrum of the emitted radiation is therefore a comb of harmonics separated by $2\omega_0$. As we demonstrated in the theoretical part (2.1.1), even-order nonlinear processes cancel out in a centrosymmetric medium. Therefore, only the odd harmonics of the laser field are emitted. A typical

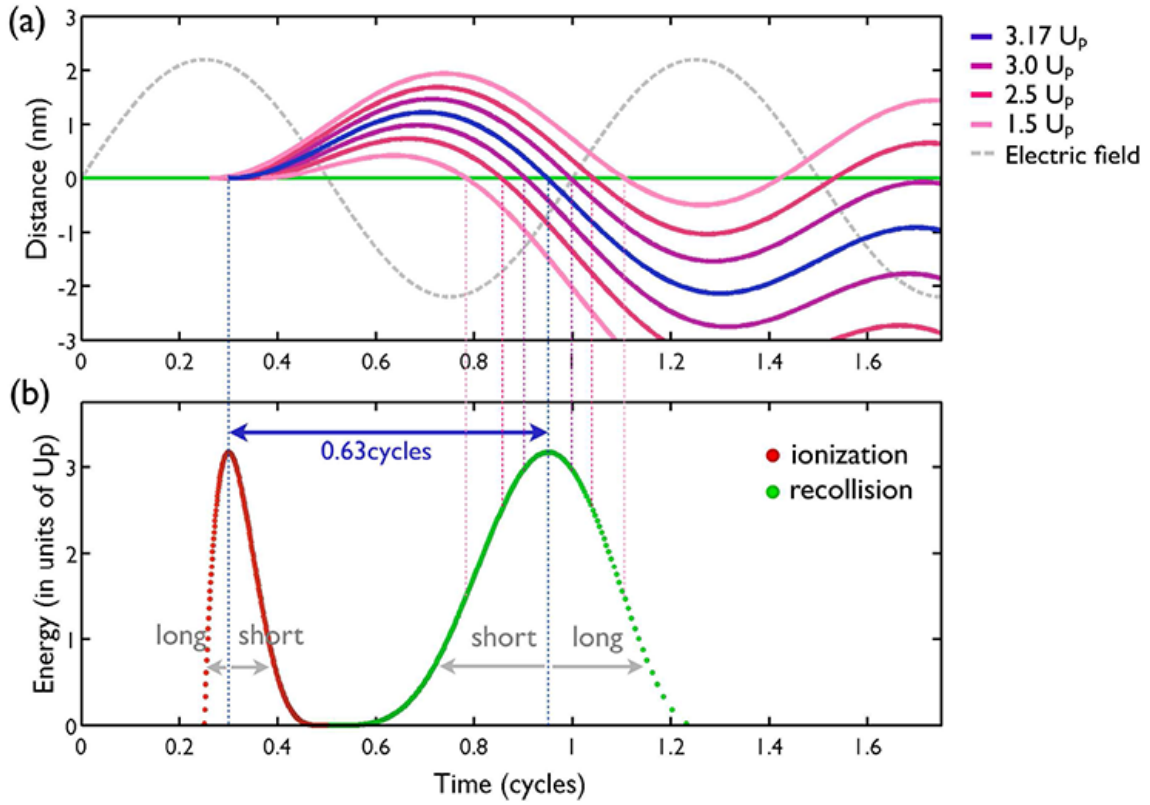


Figure 7: Fig.(a): Classical trajectories of electrons in a monochromatic laser field, dependent on the ionization time. The vertical axis represents the distance from the nucleus, with the green line indicating the ionic core's position. The laser field has a wavelength of $\lambda_0 = 800\text{nm}$ and a peak intensity of $1.56 \times 10^{14}\text{W/cm}^2$. The electric field is shown as a gray-dashed line. Three pairs of short and long trajectories are shown for recollision energies of $3.0U_p$ (purple), $2.5U_p$ (dark pink), and $1.5U_p$ (light pink). The most energetic trajectory, reaching $3.17U_p$ (blue) at recollision. Fig.(b) represents Kinetic energy at recollision as a function of ionization time (red points) and recombination time (green points).

Figure from [23]

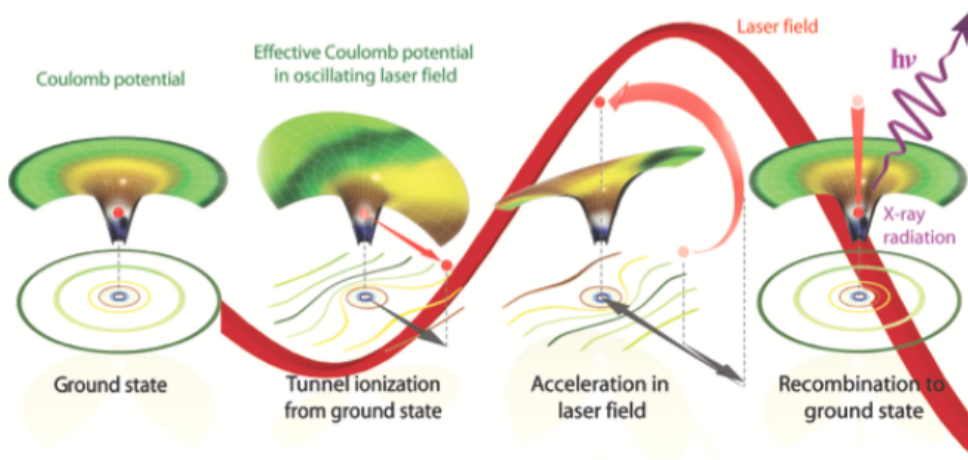


Figure 8: Illustration of the three-step model in HHG: The intense laser field distorts the Coulomb potential, causing tunnel ionization of the electron. The electron is then accelerated by the laser field before recombining with the parent ion, resulting in the emission of a photon. Adapted from [24]

high harmonic spectrum exhibits an extended plateau with near-constant harmonic intensity, and there is a clear cutoff at which harmonic intensity drops quickly as represented in Fig- 9a. Fig- 9b illustrates how the plateau and the cut-off are seen as the recombination time changes

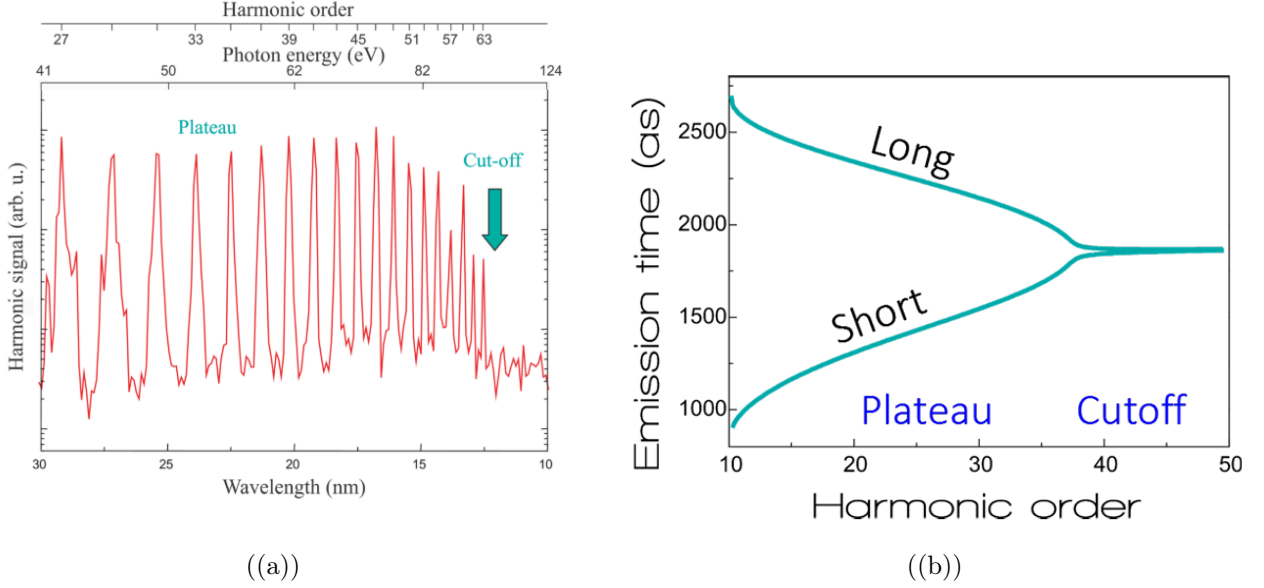


Figure 9: (a) High harmonic generation in neon gas. In this experiment, 50 fs, 800 nm pulses irradiated a sample of neon, resulting in a high harmonic spectrum. Taken from [25], (b) Correspondance between recombination time and energy at this time [19]

The three-step model does not fully capture the High Harmonic Generation process. It describes the tunneling of the electron from a quantum perspective and its propagation in the field in a classical manner. This model provides access to important parameters such as cutoff energy, ionization time, and recombination time. However, for more insights into the HHG process, for instance, the probability of recombination, the rate of ionization, etc, it is important to compare these results with those obtained from a quantum model, developed by Lewenstein in 1994. [26]

2.2.2 Temporal Structure of the Harmonic Spectrum.

The studies conducted show the existence of attosecond pulses. During radiative recombination, the electron recombines with the nucleus over a sub-femtosecond duration for each family of trajectories. In the spectral domain, the emitted radiation is sufficiently wide band (up to a few hundred eV) to allow the pulses of attosecond duration. This temporal structure of XUV light is linked to its spectral phase. The two timescales to consider here is the attosecond structure of the pulse train generated and the femtosecond structure of the envelope of the very train. They are respectively associated with the relative phase between harmonics and to the individual spectral phase of each harmonics [27]. Consider the superposition of N monochromatic harmonics, defined by their spectral amplitude A_q and the spectral phase ϕ . This would imply that we ignore the finite duration of the generating laser. The resulting time profile obtained is [27]:

$$I(t) = \left| \sum_{q=1}^N A_q e(-i\omega_q t + i\phi_q) \right|^2 \quad (55)$$

here q denotes the harmonic order. We study the behavior of this profile for different phase relationships between harmonics.

- When ϕ_q is constant, the pulse is Fourier limited, the time profile of N harmonics in phase is made up of attosecond pulses with a duration inversely proportional to N

- When ϕ_q is linear with respect to ω_q , the the temporal profile in equation 55 is shifted temporally by emission time $t_e = \frac{\partial \phi_q}{\partial \omega}$
- If ϕ_q is nonlinear, then there is a group delay associated with each frequency ω_q . The emission instance depends on the harmonic order, that is to say, the harmonics are not synchronized on the attosecond scale. The temporal profile cannot be intuitive and depends on the exact profile of the relative phases. Thus if the relative phase are random then one can have continuous emission of XUV.

3 Experimental realization in Post compression and HHG

3.1 Post compression in Multi-Pass Cell

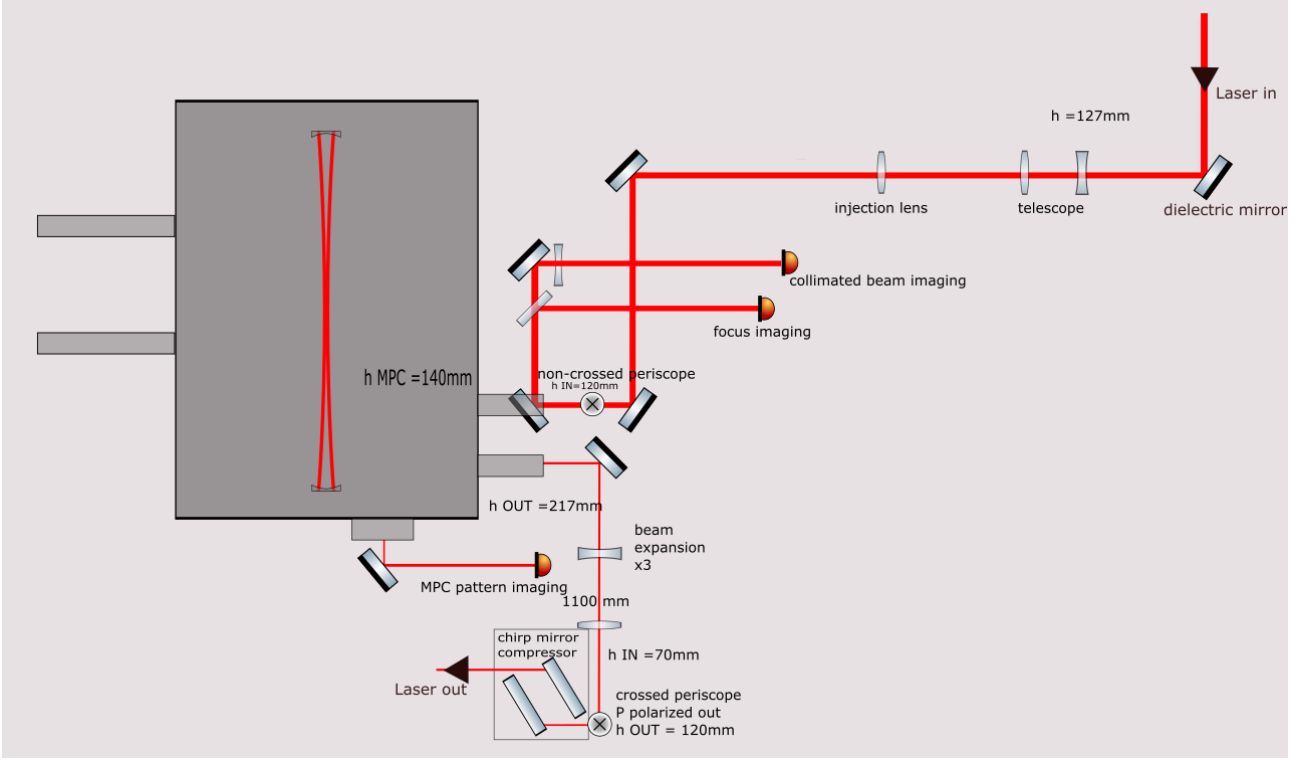


Figure 10: Beamline for spectral broadening of the pulse.

Our beamline uses a water-cooled Carbide laser from the Light Conversion company. The gain medium of the laser is a Yb: KGW crystal, and it provides pulses with a duration of 330 fs. This laser operates at a maximum output power of 80 W and a peak pulse energy of 2 mJ, with a central wavelength of 1030 ± 10 nm.

The laser beam first passes through a dielectric mirror, which reflects a wavelength of around 960-1100 nm, with 99.9 % reflectivity. This directs the beam into a telescope system for beam collimation. The telescope consists of a concave and a convex lens which adjusts the beam's diameter and divergence, ensuring a parallel beam profile over a distance.

Following collimation, the beam is focused by an injection lens with a focal length of 1500 mm, resulting in a high-intensity beam inside the multi-pass cell (MPC). Before entering the MPC, the beam traverses a periscope composed of two vertically placed mirrors. It is used for changing the beam path both and elevation. This non-crossed periscope maintains the same polarization of the beam, thus ensuring S polarization inside the MPC. The vacuum chamber is shown in Fig - 11. Leakage from the bottom mirror of the periscope is reflected by an additional mirror into a beam splitter. The beam splitter directs a portion of the beam toward two Basler cameras. One camera captures the image of the laser focus, while the other images the collimated beam after the focusing beam has been compensated with a concave lens.

The S-polarized beam exiting from the top mirror of the periscope enters the MPC vacuum chamber, which isolates its optical components from outside environment pressure and is filled with argon gas. The gas pressure inside the chamber is maintained with a manometer, with the chamber capable of holding a maximum pressure of 4 bar. Inside the MPC, the beam undergoes 27 passes and nonlinear interactions. The pattern of the beam within the MPC is imaged from the leakage of which passes through one of the cavity mirrors, which passes through a diffusing plate, itself images onto a camera. (See Fig-12)

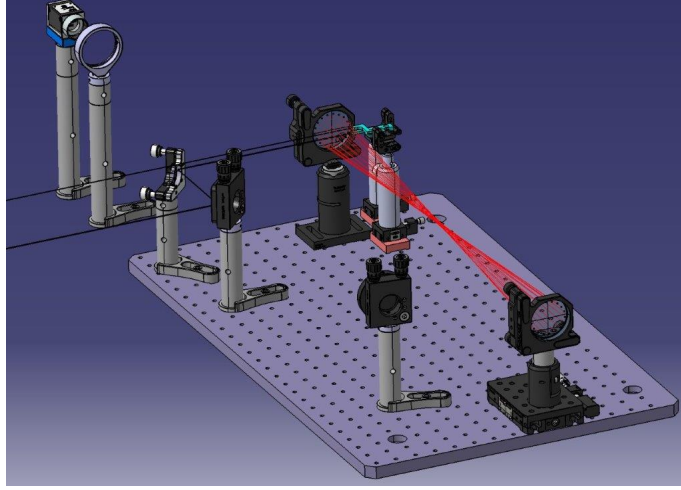


Figure 11: The vacuum chamber employed for post-compression in AttoLAB.[28]

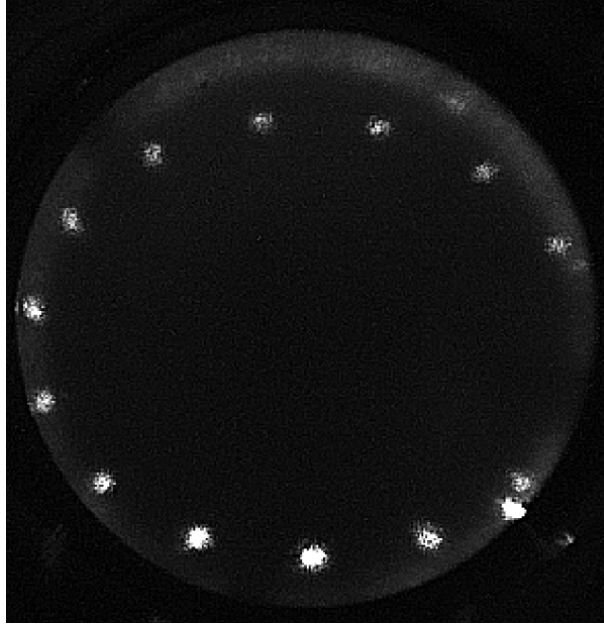


Figure 12: Reflections on the 2-inch cavity mirror.

After exiting the MPC, the beam is directed to a dielectric mirror and then propagates through a telescope, resulting in beam expansion. The beam subsequently passes through a crossed periscope, which is used to change the light's polarization from S to P. Finally, the beam enters the chirped mirror setup, undergoing 10 passes. These are dielectric multilayer mirrors where different wavelengths experience different optical path lengths. Its principle is based on Bragg mirrors, made of alternating low and high index layer, which has variable thicknesses (See Fig.13). Here we use two rectangular mirrors which allow multiple reflections, in order to tune out the dispersion introduced in the chamber.

Now we see the pressure dependence on the nonlinear refractive index. We know that n_2 is proportional to $\chi^{(3)}$. The Lorentz-Lorenz equation relates the macroscopic refractive index of a substance to the microscopic polarizability. It is expressed as [31]:

$$\frac{n^2 - 1}{n^2 + 2} = \frac{4\pi}{3} N \alpha_m \quad (56)$$

where n is the linear refractive index, N is the number density of the molecules, α_m is the mean molecular polarizability [$C.m^2/V$]. This equation has validity for homogenous solids, liquids, and gases. When the square of the refractive index is $n^2 \approx 1$, which shows that light travels similarly to in a vacuum, the

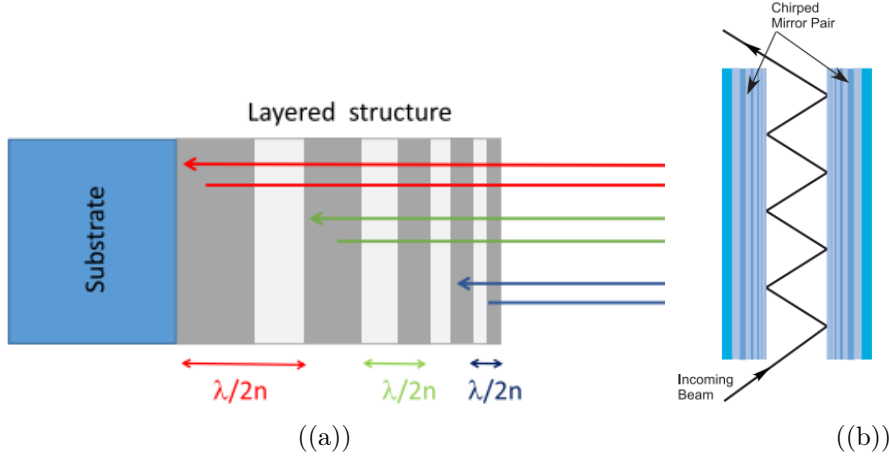


Figure 13: (a) Principle of a chirped mirror compressor. The layered structure leads to reflection of wavelength from different positions, leading to the pulse acquiring a negative chirp [29] (b) Schematics of a chirped mirror pair with 8 passes [30].

equation simplifies to $n^2 - 1 \approx 4\pi N\alpha_m$, or equivalently,

$$n - 1 \approx 2\pi N\alpha_m \quad (57)$$

For a given volume of gas, multiplying by the density of particles (pressure) allows us to go from the microscopic notion of polarizability to the macroscopic notion of the index of refraction. By extending this reasoning to the nonlinear index, we can conclude that the nonlinear index is proportional to the gas density, and thus it increases with the pressure. Consequently, the nonlinear phase accumulated will also increase as the pressure increases.

Experiments were conducted with varying pressures of argon gas inside the multipass cell (MPC) chamber and for three different pulse energies, with 1 kHz repetition rate. Given that self-phase modulation (SPM) is intensity-dependent, coming from the dependence of the refractive index on intensity, it also shows pressure dependence due to the index's dependence on pressure. This can be seen in the Fig. 14

The spectral broadening for increasing pressure and energy was observed which was as expected. After the analysis of the spectrum, we retrieve the FWHM, which is Δ or the broadening of wavelength.

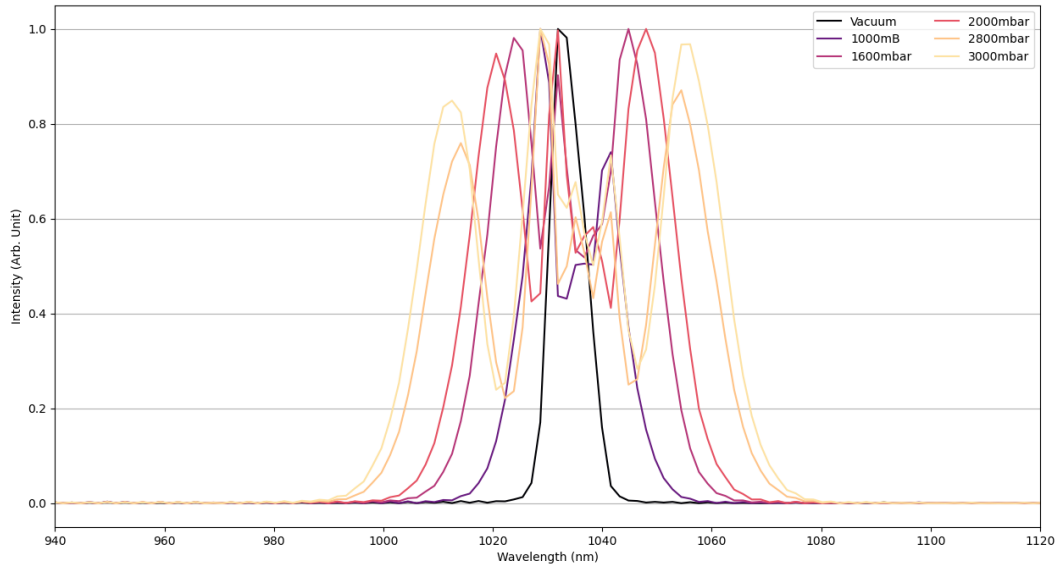
- $400 \mu J : \Delta = 60 \text{ nm}$
- $600 \mu J : \Delta = 90 \text{ nm}$
- $800 \mu J : \Delta = 128 \text{ nm}$

Fig.15 shows the FWHM retrieved for varying pressure and a fixed energy of $800 \mu J$

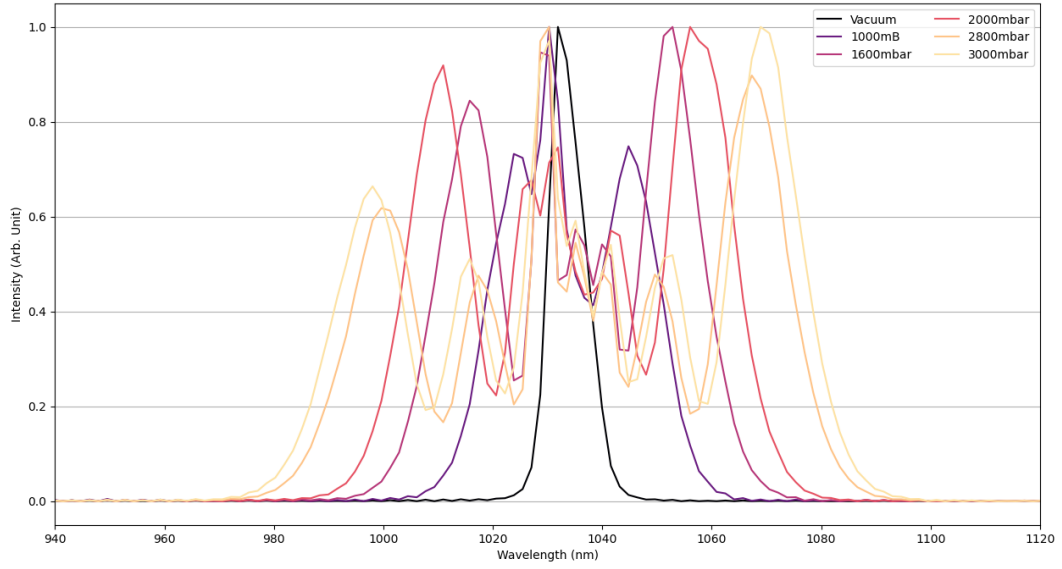
The spectrum is normalized by first correcting for background noise. The noise level is estimated by averaging the intensity values in the wavelength range where there is no signal (in this case, wavelengths less than 960 nm). This average noise level is then subtracted from all intensity values to reduce the background noise effects. Subsequently, the corrected intensity values are normalized by dividing by the maximum value in the corrected intensity data. This ensures that all the spectra are comparable in terms of relative intensity.

As mentioned before, some spectral phases will accumulate during the propagation in the MPC chamber. This is most commonly described through the concepts of Chromatic dispersion and Group Delay Dispersion.

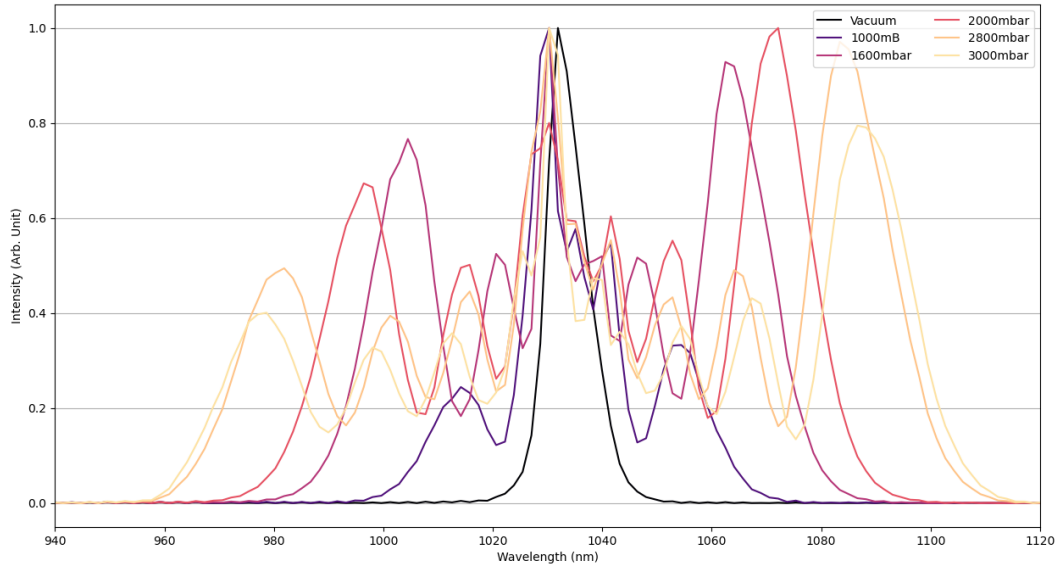
Chromatic dispersion is a phenomenon where the phase and group velocity of light vary with optical frequency as it propagates through a medium. Consequently, the refractive index $n(\omega)$ outside the vacuum is not constant over a frequency range. GDD of an optical element is a measure of chromatic dispersion.



((a)) Spectral broadening of $400\mu J$ energy per pulse



((b)) Spectral broadening of $600\mu J$ energy per pulse



((c)) Spectral broadening of $800\mu J$ energy per pulse

Figure 14: Qualitative graph of spectral broadening which shows its dependence on energy and gas pressure.

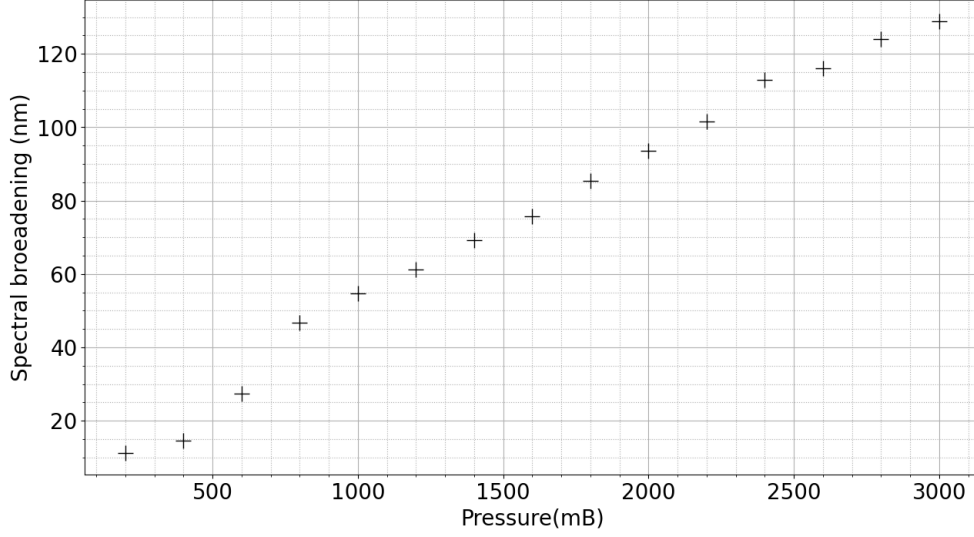


Figure 15: Spectral broadening as a function of varying pressure for 800 μJ

Sellmeier's formula can express the index variation: [15]

$$n^2(\omega) = 1 + \sum_j \frac{B_j \omega_j^2}{\omega_j^2 - \omega^2} \quad (58)$$

where ω_j is the resonant frequency of the medium, and B_j are experimentally determined fit coefficients. A pulse travelling in this medium over a distance z thus accumulates a spectral phase[32]:

$$\phi(z, \omega) = \phi(0, \omega) + k(\omega)z \quad (59)$$

where $k(\omega) = n(\omega)\omega/c$, which is the wave vector amplitude. We define the group delay given as:

$$\tau_g(z, \omega) = \frac{\partial \phi(z, \omega)}{\partial \omega} \quad (60)$$

We can define the group velocity $V_g(\omega) = 1/k'(\omega)$ which describes the velocity of the pulse envelope, and phase velocity, $V_\phi(\omega) = \omega/k(\omega)$ which is the velocity of the carrier. Writing the wave vector in the Taylor series, we get,

$$\tau_g(z, \omega) = \tau_g(0, \omega) + k'(\omega_0)z + (\omega - \omega_0)k''(\omega_0)z + \dots \quad (61)$$

Here the first term and the second term do not change the temporal envelope of the pulse. The second-order term makes different spectral components transverse with different speeds. This term is the Group Delay Dispersion (GDD), a quantitative measure of the dispersion present in an optical element. It is the second derivative of the spectral phase with respect to frequency. This is expressed as[19]:

$$GDD = \frac{\partial \tau_g}{\partial \omega} = \frac{\partial^2 \phi}{\partial \omega^2} \quad (62)$$

As observed, the spectrum broadens with increasing energy per pulse. At lower energy per pulse, we have less intensity, and the intensity-dependent refractive index decreases, and thus we have lower nonlinear phase shift, consequently less SPM, and we have lower broadening.

However, as the pressure increases, the nonlinear effects also increase. Self-Phase Modulation (SPM), which is pressure-dependent, becomes more important, as there is a change in the spectral change, leading to greater spectral broadening.

3.1.1 Measuring short pulses: The FROG technique

To characterize the pulse after the compression in the MPC and compensation through the chirped mirror compressor, we make use of Frequency-Resolved Optical Grating (FROG) measurement, specifically based on Second Harmonic Generation (SHG). It can measure the pulse duration and retrieve the spectral phase, and thus our spectrum $E(t)$. FROG works by measuring a spectrogram of the laser pulse. This is achieved by splitting the pulse into two using a beam splitter and then using one of them to gate the other through a nonlinear optical interaction like second harmonic generation, followed by a phase-retrieval algorithm [33].

To obtain measurements in both the time and frequency domain, we need both temporal and frequency resolution simultaneously. This simultaneous measurement can be done through a spectrogram, which can be expressed as:

$$I(\omega, \tau) = \left| \int_{-\infty}^{\infty} E(t)g(t - \tau)\exp(-i\omega t)dt \right|^2 \quad (63)$$

where $g(t - \tau)$ is called the gate function. Thus it is a set of spectra of all temporal gated $E(t)$ as the delay is varied. In principle, it is best to use a gate that is shorter than the pulses, however since no event is shorter than the pulse, we use pulse to gate with itself! Thus we take $g(t - \tau) = |E(t - \tau)|^2$. In our specific case of SHG FROG, we write $I(\omega, \tau) = I_{FROG}^{SHG}(\omega, \tau)$. This is analytically an impossible problem to solve. Thus FROG resorts to a numerical procedure wherein for a guess pulse, we calculate a simulated $I_{FROG}^{SHG}(\omega, \tau)$. Then, the error compared to the measured $I_{FROG}^{SHG}(\omega, \tau)$ is calculated, from which another pulse is guessed. Then, in an iterative non-linear optimization procedure, this process is repeated until the error is minimized. A typical FROG setup is illustrated in the Fig. 17

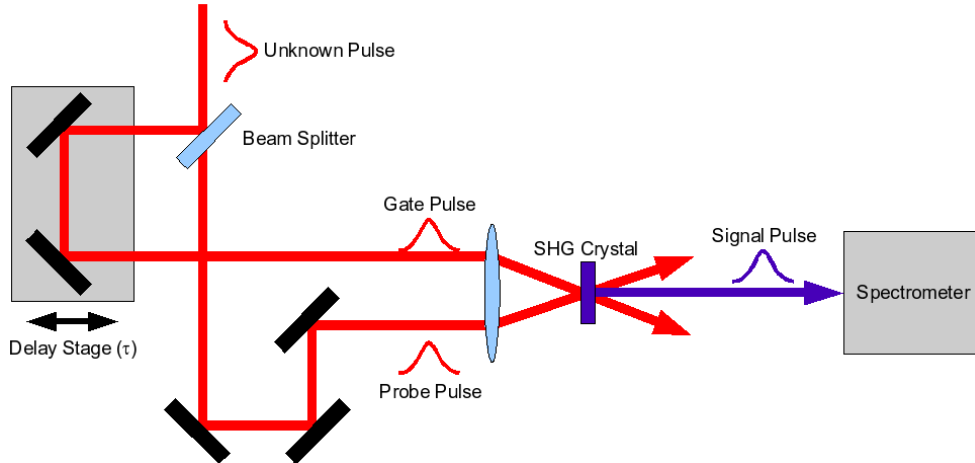


Figure 16: SHG FROG setup, where the $I_{FROG}^{SHG}(\omega, \tau)$ is can be measured. [34]

While other types of FROG measurements exist (for instead some are based on third-order $\chi^{(3)}$ processes), SHG-FROG has the advantage of being only a second-order process, which yield more signal and more sensitivity. One disadvantage of the SHG FROG is that the pulse $E(t)$ and its time-reversed replica $E(-t)$ yield the same trace, which we call ambiguity in the direction of time, although it can be removed by placing a piece of glass in the beam.[33]

The images from a FROG measurement, which in our case is a commercial device (from the FemtoEasy company), can be seen in Fig. 17. It shows the resolved SHG spectrum as a function of time delay.

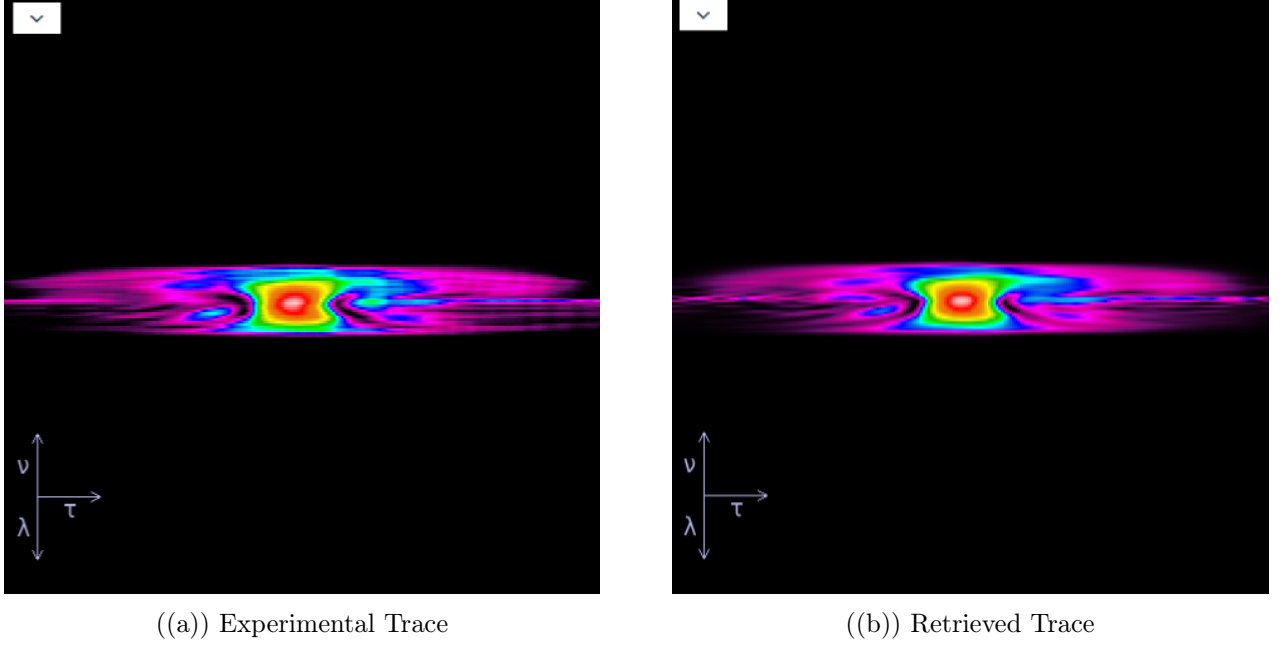


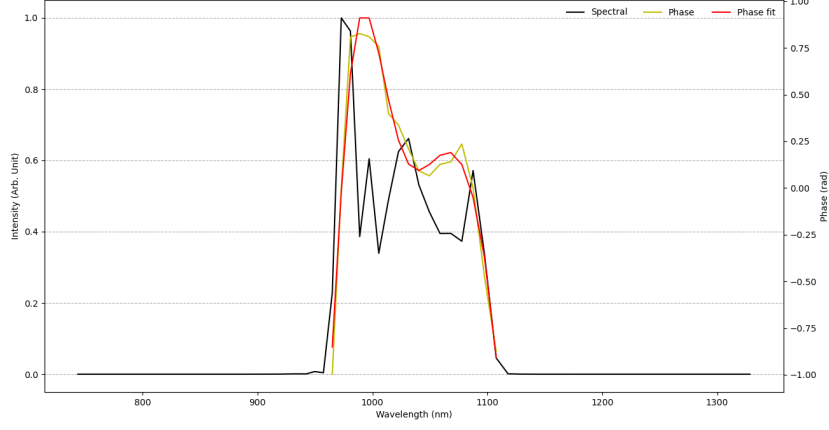
Figure 17: FROG trace, the horizontal axis is the time delay between the field, and the vertical axis is the frequency

The reconstructed spectral profile, spectral phase, temporal profile, and temporal phase, are shown in Fig.18. It can be observed that in the spectral domain, the phase is not flat: a polynomial fit (red line) shows that it has mostly third-order dispersion.

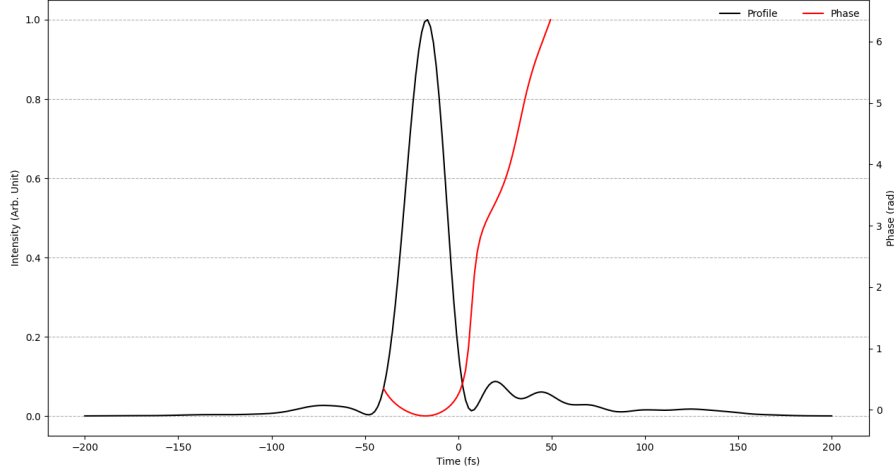
In the temporal profile, we see a Gaussian pulse with a bit of asymmetry on the right-hand side due to the third-order dispersion, and this is also reflected in the retrieved spectrum of FROG in Fig. 17, with time delay plotted on the x-axis and the frequency is on the y-axis. In the case of a Fourier-limited Gaussian spectrum, the trace looks like a uniform ellipse. Here the curvature on the side of the trace shows evidence of the third-order effect. At the center of the trace, we have the highest intensity, which is at the zero time delay.

These FROG measurements were conducted under varying pressure and energy per pulse to analyze the spectral broadening. From these measurements, we could determine the temporal compression and compare it to the Fourier-limited value, which is obtained when assuming a flat spectral phase. The results of these measurements are illustrated in Fig.19. In the optimal condition of 800 μJ and 2.8 bar, we achieved a pulse duration of 23.5 fs and a Fourier limit value of 21.7 fs.

The pulse duration is highly dependent on the spectral phase compensation applied by the chirped mirrors. As the pressure is increased to 3 bar, the MPC dispersion becomes larger than the one provided by the chirped mirror, moving away from the optimum.



((a)) Spectral profile and spectral phase



((b)) Temporal profile and temporal phase

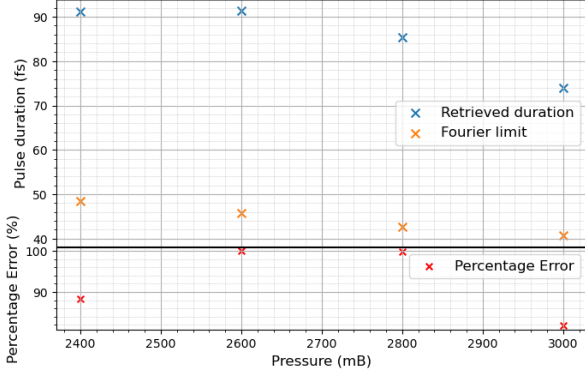
Figure 18: Retrievals from FROG for 800 μJ energy per pulse after the compression and compensation

3.2 High Harmonic Generation

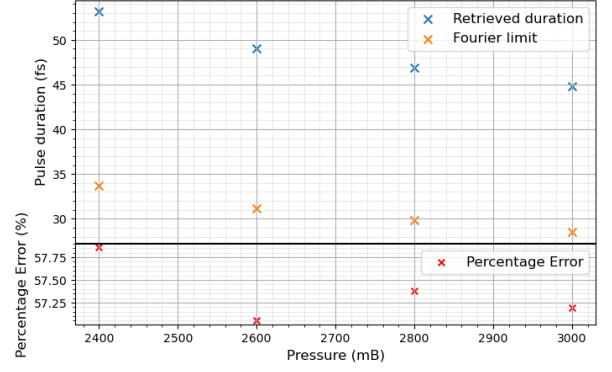
A simple scheme for high harmonic generation and its detection is illustrated in Fig. 21. For XUV light, the typical absorption length of this photon-energy spectrum is $\sim 100 \mu m$ in the air at atmospheric pressure. Therefore, the generation, propagation, and detection of the harmonics occur in a vacuum chamber. The beamline consists of a generation stage and a detection stage.

Before the vacuum chamber, the beam from the chirped mirror passes through a beam splitter and then propagates through a series of mirrors and delay stages. However, for our experiments, the delay stage and the probe beam from the beam splitter are not utilized. The beam after reflecting off the drilled mirror goes through a lens that has a focal length of 500 mm, which is mounted on an X-Y translation stage and focuses the beam into a localized gas jet inside the generation chamber. The generation chamber has a UV-fused silica window which has an anti-reflective coating for IR radiation. This would prevent the beam from reflecting and propagating back into the optical components. Inside the chamber, from the top descends an X-Y-Z manipulator which positions the gas inlet to where the beam is focused. The inlet consists of an aluminum tube, which has a tube drilled on one side, and connected to an external gas bottle on the other side. The gas is let into the chamber through this hole which is 500 μm in diameter (See Fig.20)

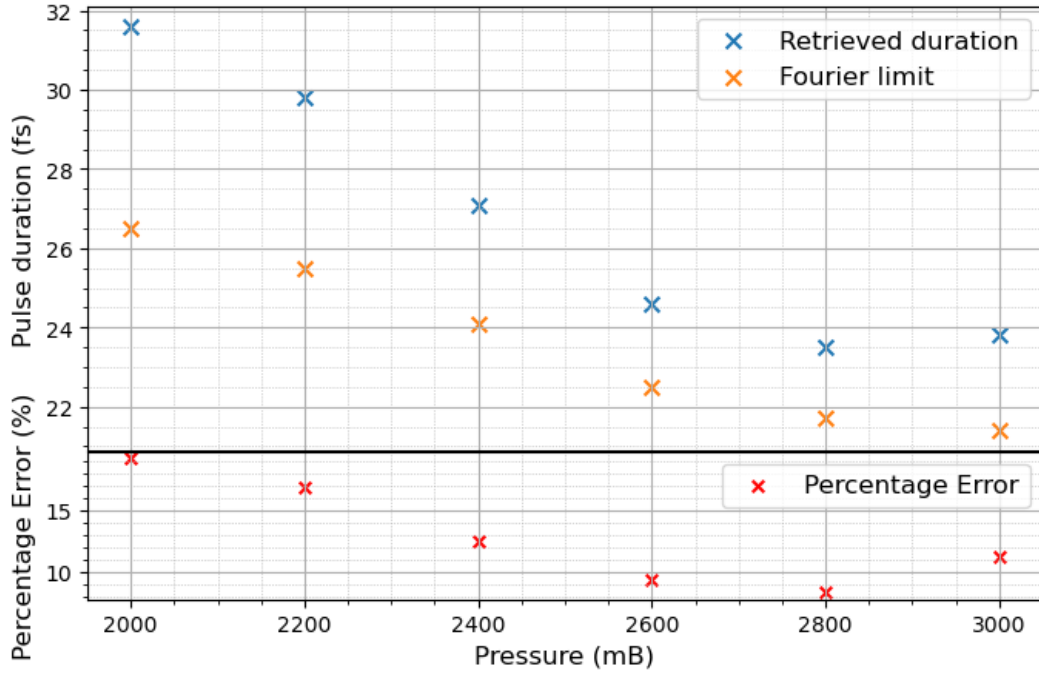
The gas expanded into the vacuum chamber has to be efficiently pumped out to ensure that the



((a)) Temporal compression of $400\mu J$ energy per pulse



((b)) Temporal compression of $600\mu J$ energy per pulse



((c)) Temporal compression of $800\mu J$ energy per pulse

Figure 19: – Nonlinear temporal compression. Top panels: FROG retrieval of the pulse duration and the associated Fourier limit. Bottom panels: deviation of the retrieval from the Fourier limit for each gas pressure, with the overcompensation for 3bar pressure.

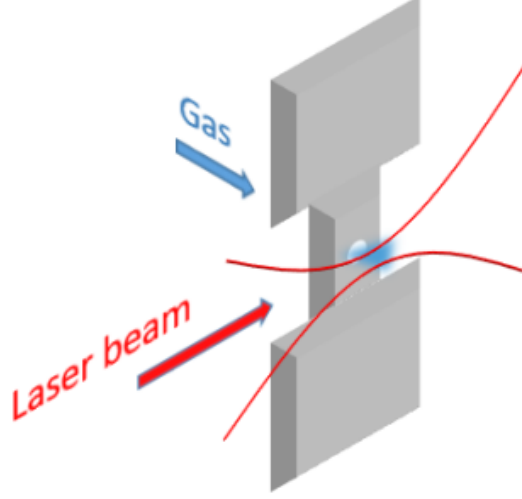


Figure 20: Illustration of the gas inlet. Figure from [35]

generated harmonics are not re-absorbed. The gas jet is positioned such that it favors the short trajectory for optimization. This can be done with the translation stage of the optical lens. The interaction of the laser beam with the gas leads to the formation of plasma. However, it must be noted that at too high intensity most of the atom gets ionized, and this prevents it from becoming the target for subsequent HHG. The plasma also leads to defocusing of the IR laser pulse, and this can reduce the laser intensity. This control of the beam size is relevant to allow for re-adjustments of intensity distribution at the focus. This can be done with an iris, which can be placed before the optical lens to aperture it down.

The detection chamber consists of an XUV photon spectrometer, and the pressure inside the chamber is maintained around 10^{-6} mbar, to house the microchannel plate (MCP) detector. The spectrometer consists of a gold-coated grating (Shimadzu, model:30-002 1623622), at grazing incidence, offering a larger collection angle, and can focus the harmonics on the detector. It is an aberration-corrected laminar-type replica diffraction grating for flat-field polychromator, where the flat field image is achieved by varying groove space. It has 1200 grooves/mm and is mounted on a translation stage. The XUV radiation propagates to a detection system, which is a stack of two microchannel plates followed by a phosphor screen anode. The electron emitted from the MCP hits the phosphor screen and emits light in the visible range. It is imaged by a CMOS digital camera (Hamamatsu Photonics, model: ORCA Spark C11440-36U), which is placed outside of this chamber. This spectrometer provides the spectrum of the emission and the spatial profiles of the harmonics in one direction. It thus allows adjusting the generating conditions for optimization of the HHG process and to yield close to Gaussian intensity profiles. The optical table with all the components used in the AttoLAB for HHG is shown in Fig: 22

To calibrate the beam size before it reaches the optical lens, we placed an iris before the lens and checked the power transmitted as the size of the aperture was increased. Assuming that the electric field is a Gaussian, we have

$$E \propto e^{-\left(\frac{r}{w_0}\right)^2} \quad (64)$$

and we can define the power as:

$$P \propto \int_0^R \int_0^{2\pi} r E^2 dr d\theta \quad (65)$$

thus we can fit the data into the Gaussian modeled as

$$P(R) = A(1 - e^{-2\left(\frac{R}{w_0}\right)^2}) \quad (66)$$

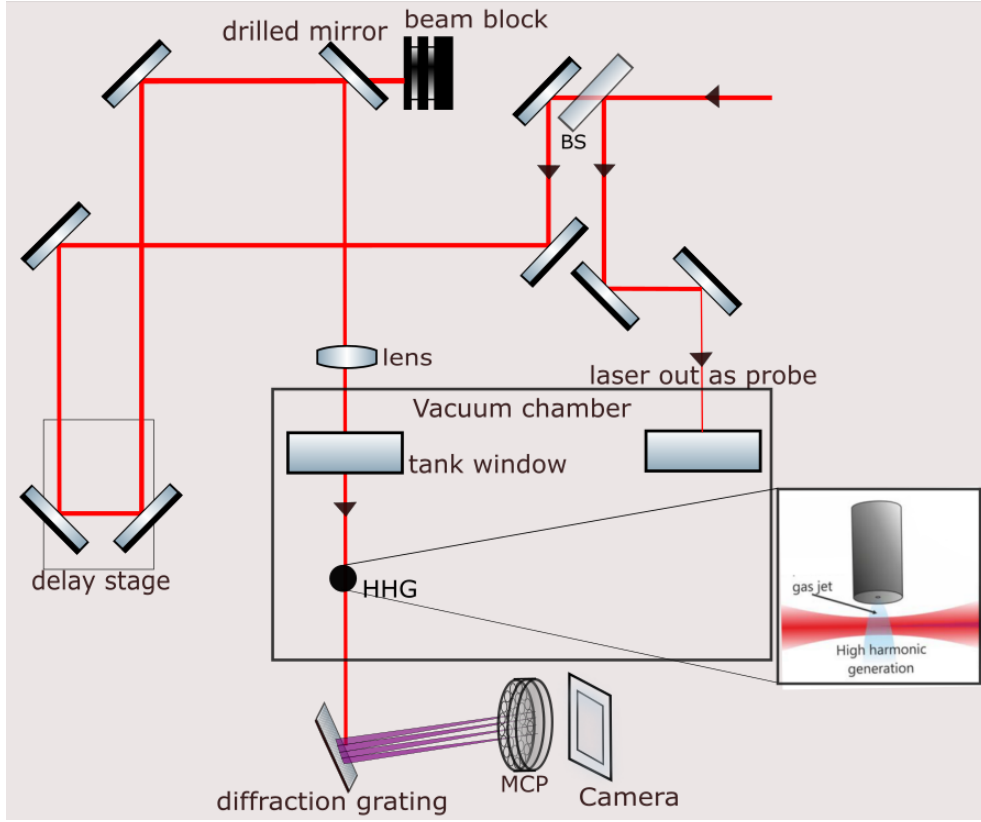


Figure 21: Beamline for HHG in FAB10

where A is a constant, w_0 is the waist of the Gaussian beam, and R is the size of the aperture. The analysis performed is in Fig. 23. The beam diameter obtained, before focusing and after the fit was 10.26 mm.

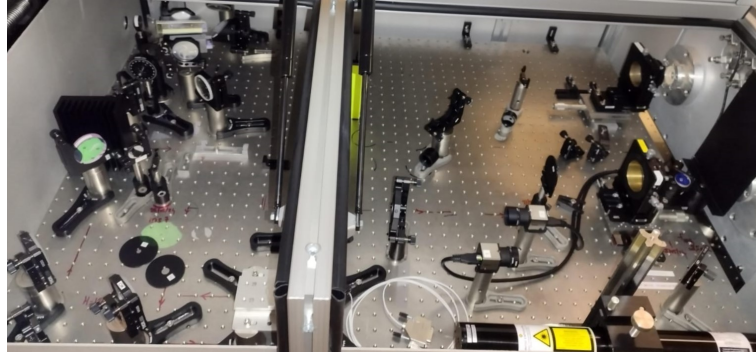


Figure 22: Experimental setup in AttoLAB.

The spatial profile obtained in the spectrometer for HHG is shown in Fig. 24. The horizontal axis corresponds to the energy dimension (increasing from left to right) and the vertical axis is the divergence. As can be seen, there is a high-intensity region, corresponding to the plateau as discussed in section 2.2.1.1, and as we move toward the higher energies, the intensity decreases, representing the cut-off region. We acquired an amplified signal by increasing the voltage on the MCP.

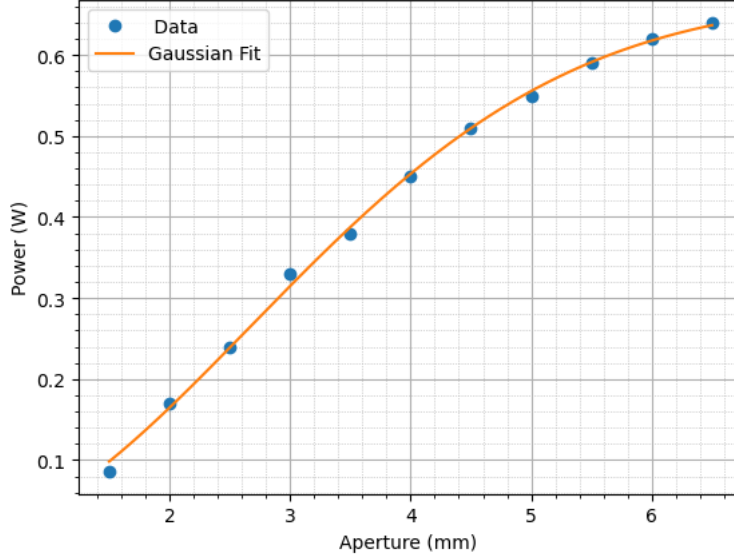


Figure 23: Power measured for varying aperture size

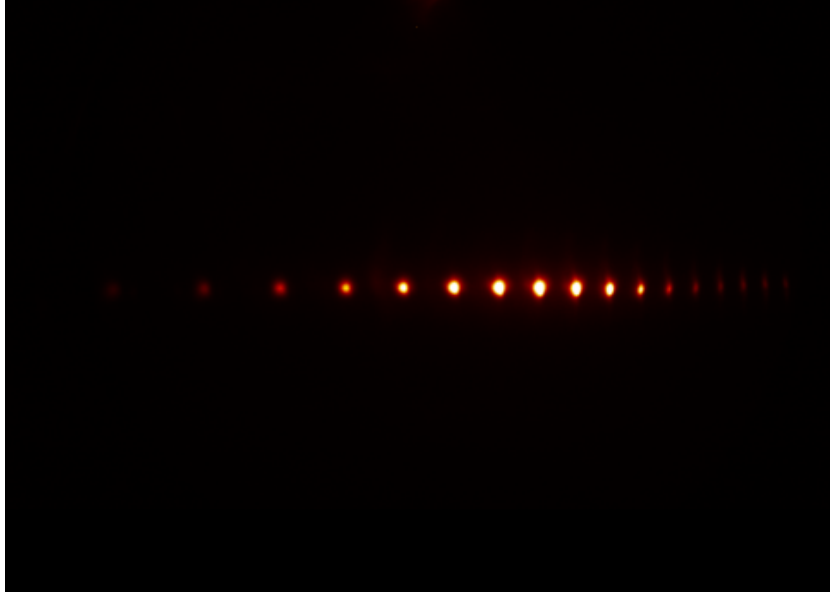


Figure 24: HHG generation in Argon, with a laser of central wavelength $\lambda = 1030$ nm, with 23 fs pulsed laser, and an applied voltage across each MCP $V = -1100V$, and $+2000V$, and the pressure inside the generation chamber is 4.86×10^{-3} mbar

An experimental aspect to be taken into account is the spacing of the harmonics, which as mentioned before, will be $2\omega_0$, theoretically, however, in the image obtained, the spacing between the lower harmonics is higher than that compared to high harmonics. This is due to the principle involved in the diffraction from a grating. When the XUV radiation hits the grating, while some light reflects directly off the grating as "0" order " diffraction, the other part of the light is diffracted based on the wavelength. The lower wavelengths are always closer to the directly reflected light from the grating. This implies that the higher harmonics would deviate less from the normal incidence, and the lower harmonics would deviate more due to the kick of the moment by the grating, which is along the surface of the grating, and this can be theorized using the equation given by,

$$\theta_i + \theta_r = \frac{n\lambda}{d} \quad (67)$$

where θ_i is the angle of incidence, θ_r is the angle of reflection, d is the distance between the grooves, and n

is the order of the diffraction, where we see a clear dependence of spacing on the wavelength. The decrease in the spacing for the higher harmonics is non-uniform since the grating equation has θ_r proportional to λ (wavelength) and λ goes as $1/E$ (energy).

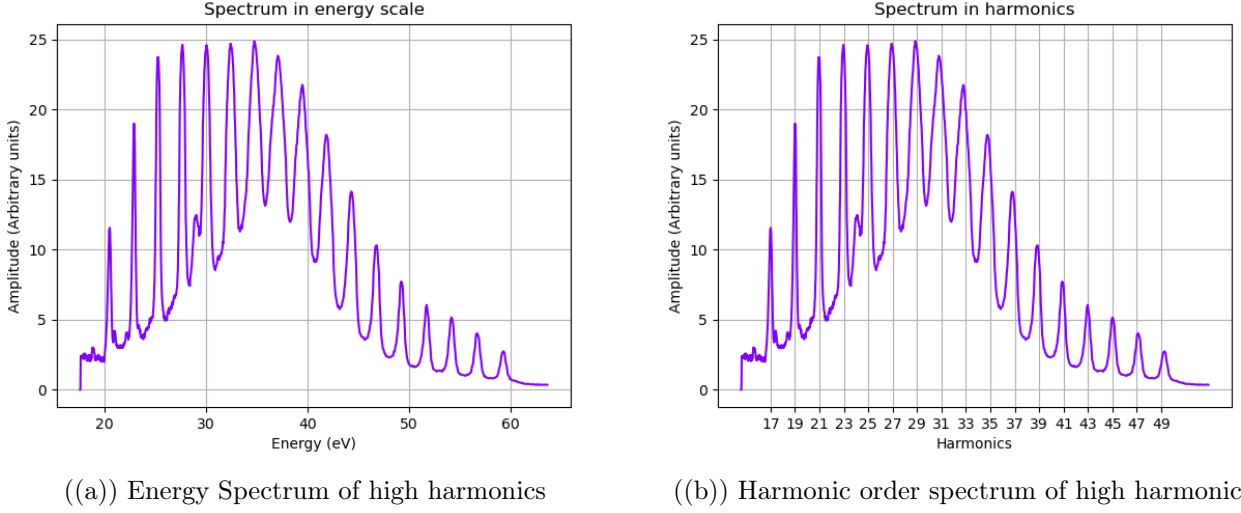


Figure 25: Spetrum obtained for the HHG in different scales

The image from the spectrometer is then used to plot the harmonic spectrum which is the Intensity vs photon energy. This can be seen in the Fig- 25. The image is calibrated using the equation 67. That is a grating fit is used to model the relationship between the harmonic number q and the pixel position on the CCD. This is converted into energy using the usual formula:

$$E_q = q \cdot E_{\text{photon}} \quad (68)$$

where $E_{\text{photon}} = 1.2$ eV is the photon energy of the driving laser field, and E_q is the energy of the given harmonics. There is an apparent background between 30eV and 40 eV, this is due to the integration of dark space on the MCP where infrared noise was present. We generated harmonic $q = 17$ to harmonic $q = 49$. This was done under the optimal energy of $800 \mu\text{J}$ and 2.8 bar pressure, with 1 kHz repetition rate. The intensity of the spectrum reduces after a particular order of the harmonic, and this is the maximum energy that can be gained by the electron during the second step of the HHG three-step process. In the region of cut-off, where both the trajectories merge, we have a degenerate trajectory with maximum energy for the photon but less generation efficiency.

4 Non-linear optics with light carrying Orbital Angular Momentum

4.1 Theoretical framework

The study of light's angular momentum and its transfer to matter can be traced back to 1992 and the seminal work of Allen and coworkers [36]. They understood for the first time that Laguerre- Gaussian (LG) modes carry a well-defined orbital angular momentum (OAM). More precisely, a LG mode propagating in the z -direction has an azimuthally varying phase, $e^{il\phi}$, carries a z -component of orbital angular momentum of $l\hbar$ per photon. This property holds in the regime of paraxial optics.

Consider a monochromatic field of light with angular frequency $\omega = kc$. The wave function of this field writes:

$$\psi(r) = u(x, y, z)e^{ikz} \quad (69)$$

with $u(x, y, z)$ the complex amplitude, and e^{ikz} is a rapidly varying phase term with $k = \frac{2\pi}{\lambda}$, being the wave number. The Helmholtz equation can be written as:

$$\nabla^2\psi + k^2\psi = 0 \quad (70)$$

substituting for the wave function in this equation, we get:

$$\nabla^2(ue^{ikz}) + k^2(ue^{ikz}) = 0 \quad (71)$$

and expanding the Laplacian term, we get:

$$(\nabla^2u)e^{ikz} + 2(\nabla u + \nabla e^{ikz}) + u\nabla^2e^{ikz} + k^2ue^{ikz} = 0 \quad (72)$$

further simplifying each term:

$$\nabla e^{ikz} = ik\hat{z}e^{ikz} \quad (73)$$

$$\nabla^2e^{ikz} = -k^2e^{ikz} \quad (74)$$

Thus the equation becomes:

$$(\nabla^2u)e^{ikz} + 2ik\frac{\partial u}{\partial z}e^{ikz} - k^2ue^{ikz} + k^2e^{ikz} = 0 \quad (75)$$

and cancelling out the common factor e^{ikz} the equation becomes:

$$\nabla^2u + 2ik\frac{\partial u}{\partial z} = 0 \quad (76)$$

In the paraxial approximation, $u(x, y, z)$ varies slowly along the direction of propagation z . This means the second derivative term is $\frac{\partial^2u}{\partial z^2}$ small as compared to the first derivative term, thus we use the approximation,

$$\frac{\partial^2u}{\partial z^2} \ll 2ik\frac{\partial u}{\partial z} \quad (77)$$

And neglecting the second derivative, we simply the Helmholtz equation to:

$$i\frac{\partial u}{\partial z} = -\frac{1}{2k}\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)u \quad (78)$$

and this equation to similar to the Schrodinger equation. To connect this equation to the field vectors, we work in Lorentz gauge condition, given by:

$$\nabla \cdot \mathbf{A} + \frac{1}{c^2} \frac{\partial \phi}{\partial t} = 0 \quad (79)$$

and write the vector potential as [37]:

$$\mathbf{A} = (\alpha \hat{\mathbf{x}} + \beta \hat{\mathbf{y}}) u e^{ikz} \quad (80)$$

Here α and β are a pair of complex numbers with $|\alpha|^2 + |\beta|^2 = 1$, and $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ are unit vector in x and y directions. The Lorentz gauge is used instead of the Coulomb gauge, given by $\nabla \cdot \mathbf{A} = 0$ which imposes that $\mathbf{A}$ should be transverse, however in this case, $\mathbf{A}$ can have longitudinal components, as long as the gauge condition is satisfied. Within paraxial approximation, the corresponding positive-frequency components of the electric and magnetic field are[38]:

$$\mathbf{E} = i\omega \mathbf{A} - \nabla \left(\frac{c^2}{i\omega} \nabla \cdot \mathbf{A} \right) \quad (81)$$

$$= \left[i\omega\alpha u \hat{\mathbf{x}} + i\omega\beta u \hat{\mathbf{y}} - c \left(\alpha \frac{\partial u}{\partial x} + \beta \frac{\partial u}{\partial y} \right) \hat{\mathbf{z}} \right] \quad (82)$$

$$\mathbf{B} = \left[i\beta k u \hat{\mathbf{x}} + i\alpha k u \hat{\mathbf{y}} + \left(\beta \frac{\partial u}{\partial x} - \alpha \frac{\partial u}{\partial y} \right) \hat{\mathbf{z}} \right] \quad (83)$$

The time average momentum density, derived from the electric and magnetic fields, is given by [39]:

$$\bar{\mathbf{p}} = \frac{\epsilon_0}{2} (\mathbf{E}^* \times \mathbf{B} + \mathbf{E} \times \mathbf{B}^*) \quad (84)$$

$$= \frac{\epsilon_0}{2} [i\omega(u \nabla u^* - u^* \nabla u) + 2\omega k |u|^2 \hat{\mathbf{z}} + \omega \sigma \nabla |u|^2 \times \hat{\mathbf{z}}] \quad (85)$$

here $\sigma = i(\alpha\beta^* - \alpha^*\beta)$. For a better understanding, we move to cylindrical polar coordinated (ρ, ϕ, z) .

In the components of this momentum density, the second term represents the linear momentum in the z propagation direction. This is consistent with the idea that each photon carries a momentum $\hbar k$ in the direction of propagation. In paraxial approximation, we can ignore the z -component of the first term.

The angular momentum, or the twisting nature of the light about its propagation direction, is associated with the ϕ component of the momentum density. Two terms can give rise to a ϕ -component : the first term, $(u \nabla u^* - u^* \nabla u)$ and the last term, $\nabla |u|^2 \times \hat{\mathbf{z}}$. From the second term, this can arise when $\alpha\beta^*$ has an imaginary part which is the measure of circular or elliptical polarization of the beam. This term is associated with the spin which is defined by electric field polarization. Fig- 26 shows an example of how a quarter wave plate can generate circularly polarized light from linearly polarized light.

Let us now consider the following ansatz for the complex amplitude:

$$u = v(\rho, z) e^{il\phi} \quad (86)$$

where $v(\rho, z)$ is a function that depends on the radial coordinate ρ , (also associated with the diffraction of light) and the axial coordinate z , and $e^{il\phi}$ is the azimuthal phase dependence, where l is an integer representing the OAM of the light beam.

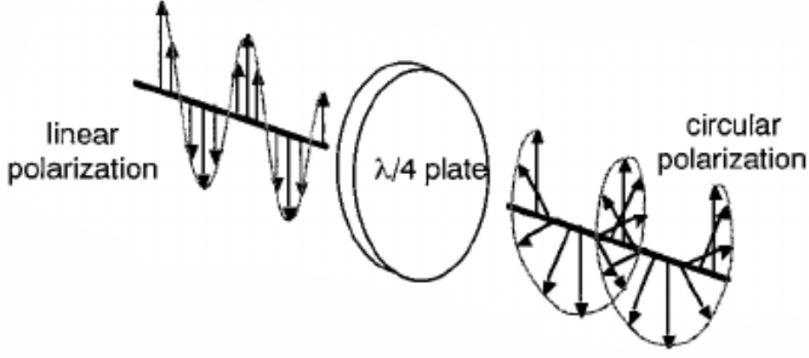


Figure 26: Spin Angular Momentum defined by the electric field polarization. The quarter wave plate changes the polarization of linearly polarized light to circularly polarized light. Adapted from [40]

For a beam with the amplitude of the form (equation:86) , the angular momentum density in the cylindrical coordinates is given by [36]:

$$j_z = \rho \bar{p}_\phi = \epsilon_0 \omega l |u|^2 - \frac{\epsilon_0}{2} \omega \sigma \rho \frac{\partial |u|^2}{\partial \rho} \quad (87)$$

where $\bar{p}_\phi$ is the ϕ component of the time- averaged momentum density $\bar{p}$. In the paraxial approximation, the flux of the angular momentum is given by $c j_z$, where c is the speed of the light. The total flux of energy is given by the Poynting vector, whose z - component is [38] :

$$\bar{p}_z = \epsilon_0 \omega k |u|^2 \quad (88)$$

thus the ratio of the total flux of angular momentum to the total flux of energy is [38]:

$$\frac{\int \int \rho d\rho d\phi c j_z}{\int \int \rho d\rho d\phi c^2 \bar{p}_z} = \frac{l + \sigma}{\omega} \quad (89)$$

Multiplying and dividing this equation by $\hbar$ we get $\frac{\hbar l + \hbar \sigma}{\hbar \omega}$. This result is interpreted as:

- Each photon is associated with an energy $\hbar \omega$
- The term $\hbar l$ represents the Orbital angular Momentum (OAM) per photon, for a wave with an amplitude having azimuthal dependence $e^{il\phi}$
- The term $\hbar \sigma$ represents the Spin Angular Momentum (SAM) per photon.

4.1.1 Phase singularities

We have established the fact that a light beam with an amplitude having azimuthal dependence, non-zero topological charge l , can carry an OAM quantized in units of $\hbar l$ per photon. Thus OAM is directly linked to the topology of wavefronts. When moving around the z -axis in a closed loop, the phase will change by

$$\oint_C \nabla \phi \cdot d\mathbf{s} = 2\pi l \quad (90)$$

where $\mathbf{s}$ is the unit vector tangential to the closed path C around which the loop is taken. The amplitude has an undefined phase or phase singularity (also known as an optical vortex, a topological defect)) on the z - axis, that is, at $\rho = 0$. Since the phase is not defined at this point, it necessarily follows that the field will have zero intensity there and we will see that this is a common feature for fields carrying orbital angular momentum. A representation of phase singularity can be seen in Fig.27

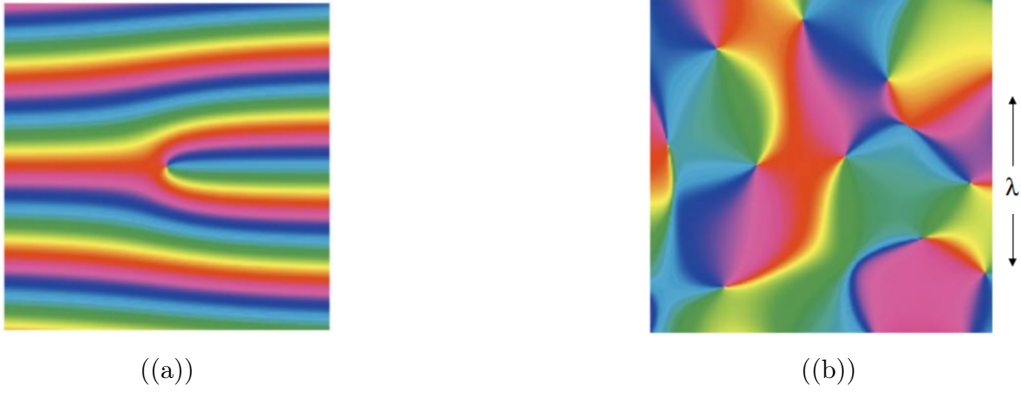


Figure 27: (a) A plane wave with a phase singularity. At this point, ϕ changes by multiples of 2π around the singularity, creating a discontinuity or a "dislocation" in the phase. (b) The superposition of plane waves contains many singularities and in non-monochromatic waves, they can move faster than light, since they carry no energy [41]

4.1.2 Laguerre- Gaussian beam

Laguerre Gaussian modes u_{pl}^{LG} are the solution to the paraxial wave equation in cylindrical coordinates. The general form of LG mode is [42]:

$$u_{pl}^{LG} = \frac{C_{pl}^{LG}}{w(z)} \left(\frac{\sqrt{2}\rho}{w(z)} \right)^{|l|} L_p^{|l|} \left(\frac{2\rho^2}{w(z)^2} \right) e^{-\frac{\rho^2}{w(z)^2}} e^{il\phi} e^{ik\frac{\rho^2 z}{2(z^2 + z_R^2)}} e^{-i(2p+|l|+1)\tan^{-1}(z/z_R)} \quad (91)$$

where $p \in N$ and $z \in z_R$ is the Rayleigh range, $w(x) = 2[(z^2 + z_R^2)/kz_R]^{1/2}$ is the radius of the beam. The focal plane of the LG mode is at $z = 0$ and has the smallest width there. $L_p^{|l|}$ is the generalized Laguerre polynomial, and is given by:

$$L_p^{|l|} = (-1)^{|l|} \frac{d^{|l|}}{dx^{|l|}} L_{p+|l|}(x) \quad (92)$$

and the constant is taken such that modes are normalized in the transverse plane $\int \int dx dy |u|^2 = 1$ and is written as:

$$C_{pl}^{LG} = \left(\frac{2p!}{\pi(p+|l|)!} \right)^{1/2} \quad (93)$$

The term $(2p + |l| + 1) \tan^{-1}(z/z_R)$ is called the Gouy phase, which characterizes the beam's phase evolution, in addition to kz along the propagation direction. For a Laguerre- Gaussian beam, Fig.28

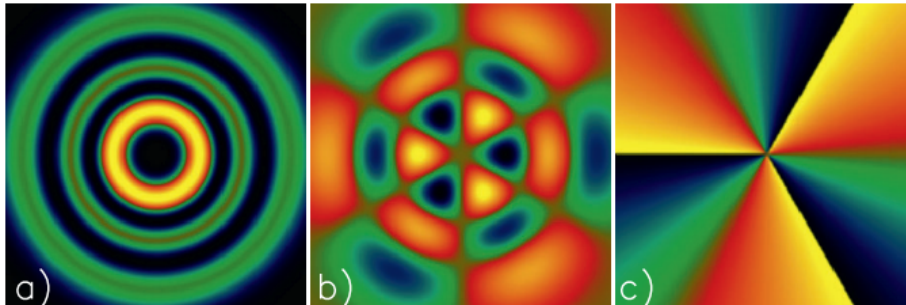


Figure 28: (a) Intensity (b) real part (c) Phase for a LG beam. Figure from [38]

shows the intensity, the real part, and phase with $l = 3$ and $p = 2$ in its focal plane.

Another solution for the paraxial equation is Bessel beams, which also carry OAM, however, this will

not be discussed in this report.

4.1.3 Generation of Light Carrying OAM

To obtain modes carrying an OAM, we need to impart an azimuthal phase dependence of $e^{il\phi}$ to a beam. One of the simplest methods to impart OAM to a light beam involves using a Spiral Phase Plate. It is a transparent material, whose thickness increases linearly with the azimuthal angle ϕ . This means that as you move around the beam in a circle, the thickness of the material steps up like a spiral staircase (See Fig.29)

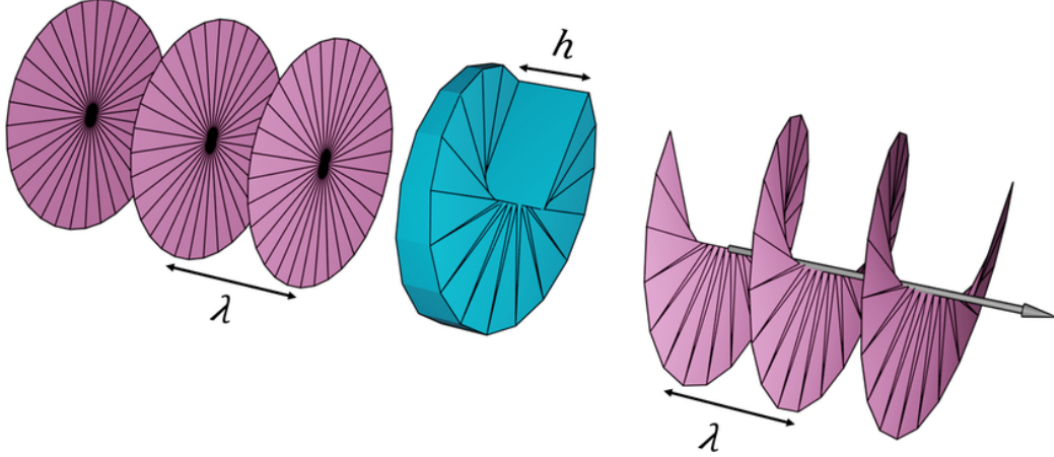


Figure 29: Spiral phase plate converting a wavefront of a plane wave to a helical wavefront of $l = 1$.
Adapter from [43]

The key idea is that this thickness variation creates a phase difference in the light passing through the material. If the total step height of the "staircase" corresponds to a phase difference of 2π (one multiple of the full wavelength), then as light passes through this material, it gets imprinted with a phase profile of $e^{il\phi}$.

When a Gaussian beam, which has a simple symmetric intensity profile and has no OAM, passes through the spiral phase plate, the beam coming out will carry $l\hbar$ units of OAM per photon. Initially, the intensity profile of the beam is unaffected, such that the beam is now a superposition of many Laguerre-Gaussian modes generated with a range of values of p and a fixed l value. However, the superposition is dominated by the $p = 0$ mode, meaning that the most significant contribution to the beam's intensity profile comes from the simplest Laguerre-Gaussian mode with no radial nodes. This $p = 0$ has a characteristic doughnut shape, which is most prominent when a Gaussian beam is transformed by a spiral phase plate to carry OAM.

4.2 Ongoing Experiments on Light Carrying OAM

The experiments conducted in the lab for the generation of a beam with Orbital Angular Momentum (OAM) follow a standard experimental setup with an additional phase plate in the beamline. The vortex plate is made of Liquid Crystal Polymer between N-BK7 glass plate. It has an outer diameter of 25.4 ± 0.2 mm. The plate has a topological charge of $l = 1$ and is designed for 1064 nm. The vortex plate is placed between the mirror and the non-crossed periscope and is mounted on an X-Y translation mount to allow precise positioning. All the measurements we conducted with 1 kHz repetition rate unless mentioned otherwise.

4.2.1 Beam profiling before post-compression

The vortex plate is placed in the beamline before the MPC, leading to the observed beam profile is shown in Fig -30. We directly see the presence of a singularity at the center of the beam profile, as expected for a vortex beam. An experimental detail that needs to be noted is the appearance of rings in the collimated beam. These rings result from one of the irises being partially closed in the beamline, leading to diffraction effects. Consequently, the beam diameter is slightly larger compared to its original, unchanged spatial profile.

To better understand how an iris can affect the beam, we can examine the focusing properties of a Gaussian beam using a converging lens. When a Gaussian beam passes through a lens, the beam waist at the new focus, denoted as ω'_0 , can be determined using the following formula [44]:

$$\omega'_0 = \frac{1}{\omega_0} \frac{\lambda f}{\pi} \quad (94)$$

where f is the focal length of the converging lens, ω_0 is the beam waist before passing through the lens. This formula highlights the inverse relationship between the beam waist before and after focusing. Thus, with a larger initial beam waist on the lens, after the focus, the beam would be even smaller.

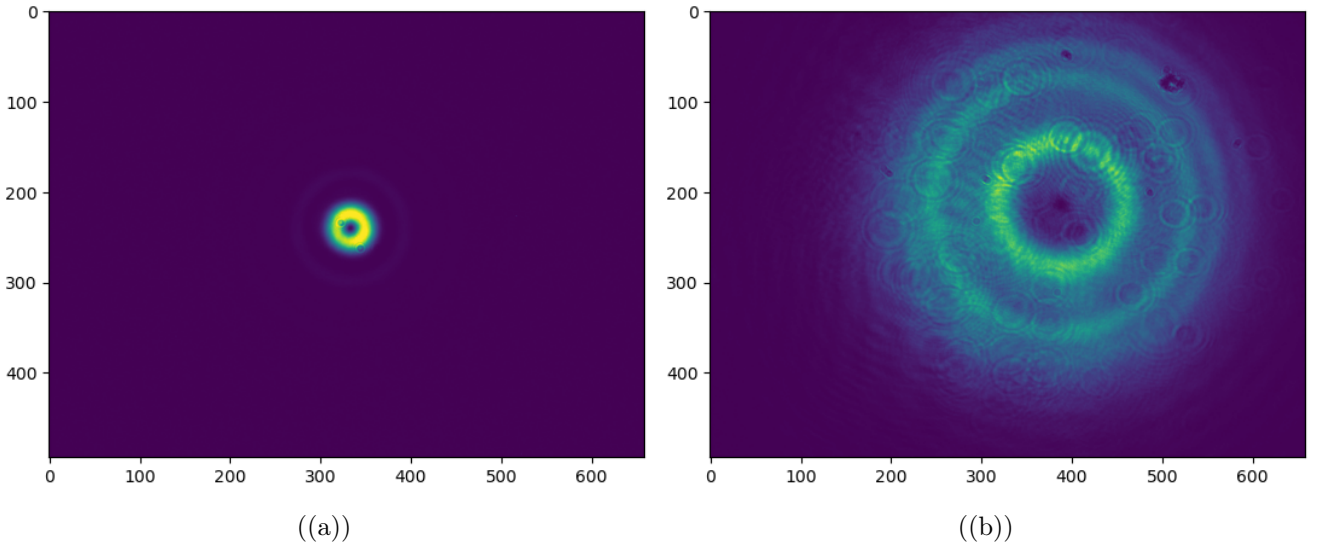


Figure 30: Beam profile of LG_{10} when (a) At focus, (b) Collimated, with the axis denoting the pixel

4.2.2 Nonlinear temporal compression of Vortex beam

After passing through the MPC, spectral broadening was observed using a spectrometer, shown in Fig-31. The pressure was 2.8 bar, as in Fig 19 which provided optimum pulse compression for the Gaussian case. The figure has been plotted with an offset along the direction of the vertical axis to prevent the overlap of the spectrum and thus is not normalized.

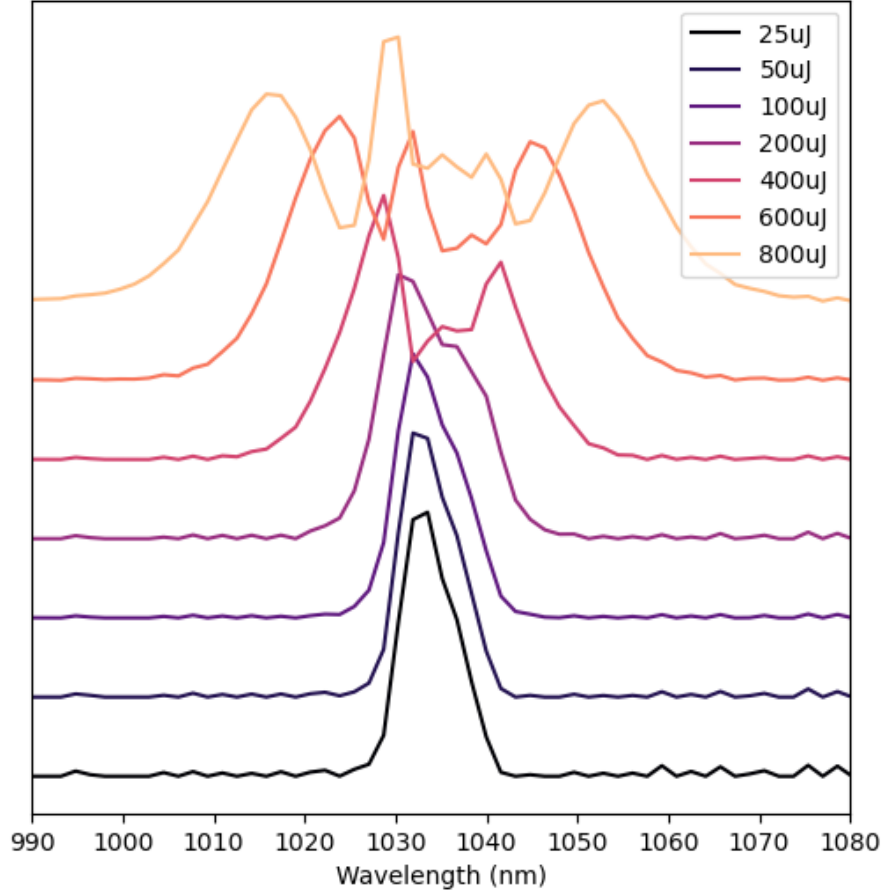


Figure 31: Spectral broadening after MPC for varying energy and 2.8 bar pressure.

It can be seen that with OAM, we still have spectral broadening, with the highest broadening obtained for $800 \mu J$. It is to be expected, as we saw a similar trend in the case without the OAM beam. For a more quantitative measurement, we find the FWHM and retrieve the plot as shown in Fig: 32.

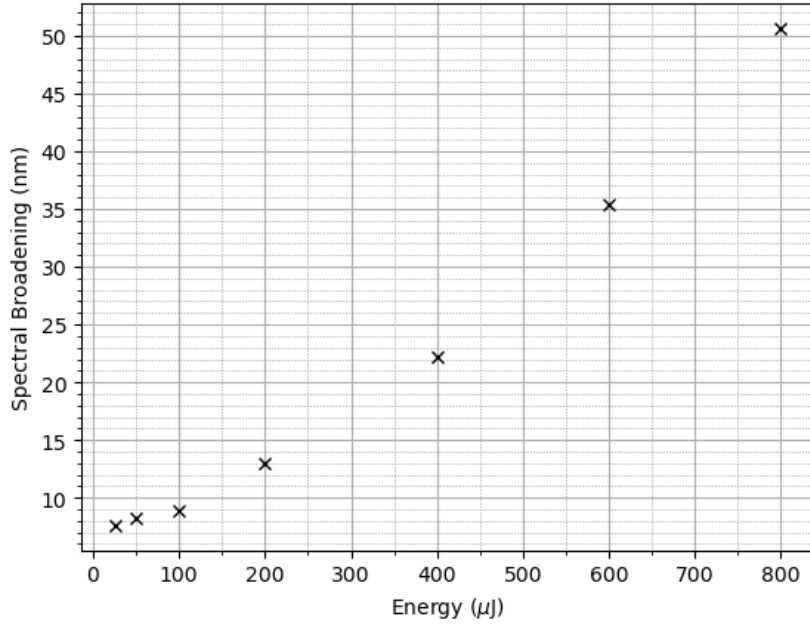


Figure 32: Quantitative measurement for spectral broadening with varying energy and constant pressure

The spectral broadening acquired for $800 \mu J$ was 50 ± 1 nm. The plot was obtained after finding the peaks of each spectrum and finding the intensity corresponding the half of the peak intensity. The data had to be interpolated on a finer spectral grid to get a more precise measurement of this quantity.

To verify the beam spatial properties in the presence of broadening, we imagine the beam profile at focus using a camera, placed after the chirped mirror compressor. The resulting images, taken at different energy levels, are shown in Fig. 33. These images were captured at a 5 kHz repetition rate of the laser with varying energy. To analyze the spatial profile of the beam, a lineout of the beam profile was taken,

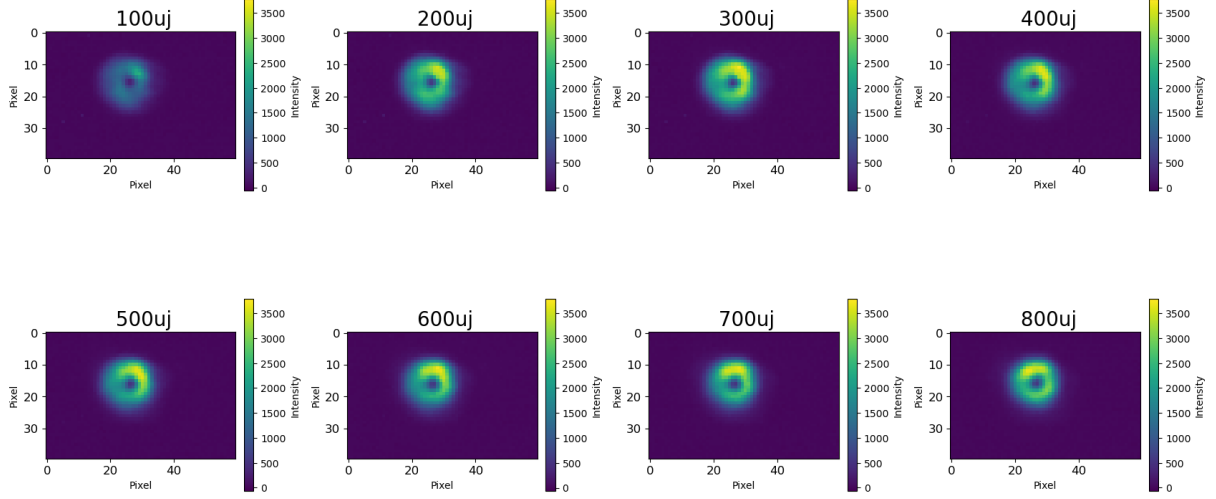


Figure 33: Variation of intensity on the beam profile with increasing energy.

as shown in Fig. 34, which shows a two-peak profile, as expected for a Laguerre Gaussian beam. The profile has been normalized by its maximum intensity for each spectrum. It is important to note that the lineout does not exhibit a uniform distribution due to experimental factors, From these images images, it can be seen that the beam retains a nice Laguerre-Gaussian profile when being broadened, suggesting that the SPM process conserves the mode of the incoming beam.

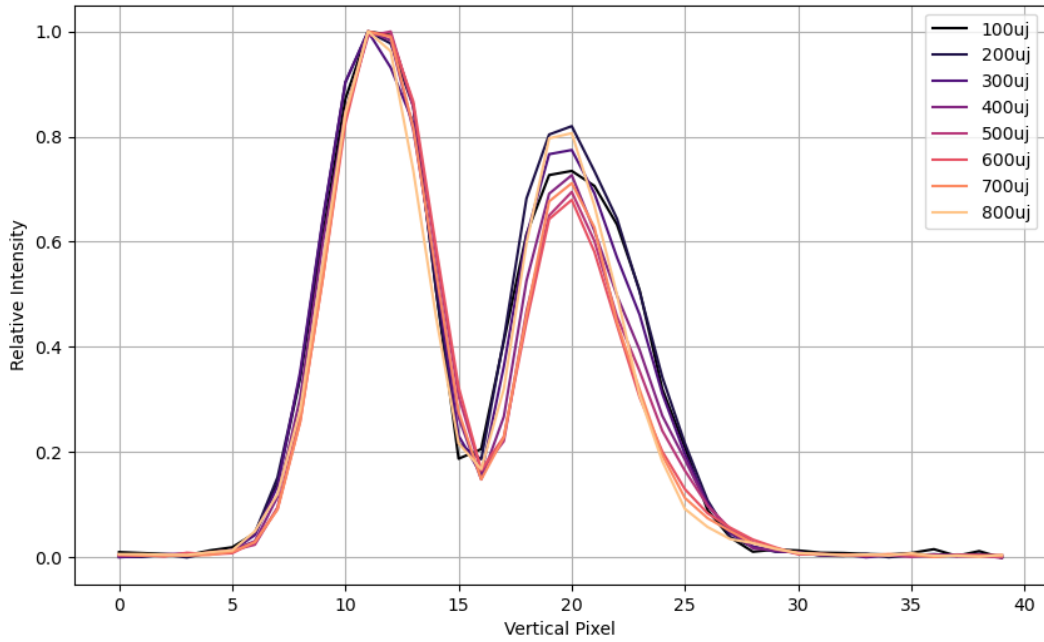


Figure 34: Lineout of the spatial profile along vertical direction

4.2.3 Vortex beam through a Triangular Aperture

In this section, we explore how the diffraction of a vortex beam through a triangular aperture produces a diffraction pattern in the far field that is related to the topological charge l . This would further confirm the conservation of orbital angular momentum during the broadening.

At a fixed plane at position $z = z_0$, the relative intensities of the diffracted field E_d is given by the integral [45],

$$E_d(\mathbf{k}_\perp) = \int \tau(r_\perp) E_i(\mathbf{r}_\perp) e^{i\mathbf{k}_\perp \cdot \mathbf{r}_\perp} d\mathbf{r}_\perp \quad (95)$$

In this equation, $E_d(\mathbf{k}_\perp)$ can be obtained from the Fourier transform of the product of the function describing the aperture $\tau(r_\perp)$ and the incident field. The transverse wave vector $\mathbf{k}_\perp$ is associated with the coordinate system of the far field region, acting as the reciprocal space.

Fig. 35 shows the integral numerically evaluated by using high order LF beam with radial index $p = 0$, as the initial condition for electric field. [46] and the diffraction patterns for different values of the topological charge of l .

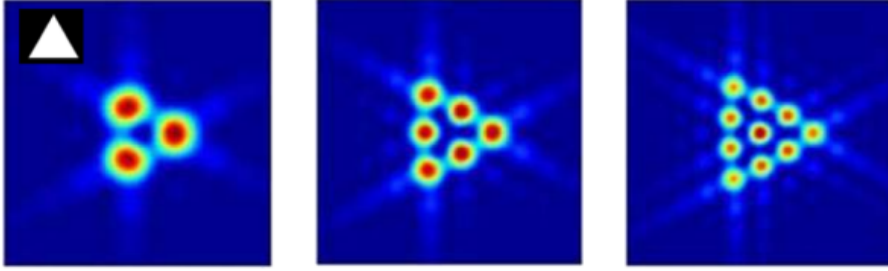


Figure 35: Numerical simulation for different topological charge l , ranging from 1 to 3. The intensity increases from red to blue.[46]

To conduct this experiment, we introduced a triangular slit into the collimated beam, positioned between the lens and the chirped mirror compressor. The resulting images are depicted in Fig .36

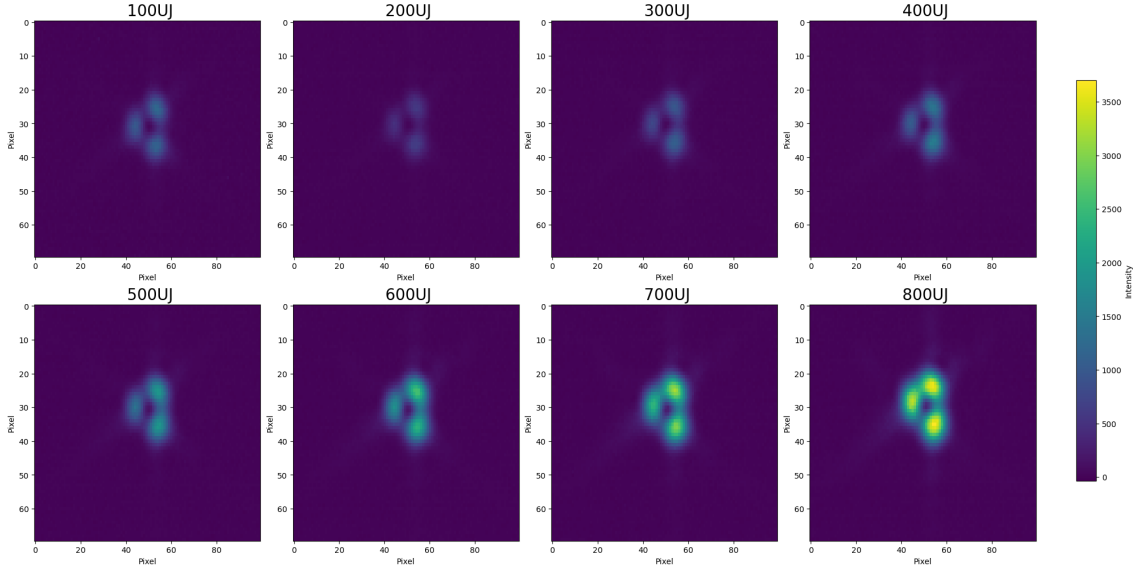


Figure 36: Dominance of $\ell = 1$, OAM with increasing energy

We see the expected three diffraction dots, which means that the beam carries an orbital angular momentum (OAM) of $\ell = 1$. The orientation of the triangular aperture placed is different from the one in Fig.35, thus in our acquisition we see that the pattern is rotated by 180° . Note that the first image, acquired with an energy of $100 \mu\text{J}$, serves primarily for qualitative measurements. The exposure time was set to auto to ensure the signal was visible, making this image non-comparable to subsequent ones that

have manual exposure conditions.

An anomaly observed here was, as the intensity there was a slight modification of the distance between the dots. A lineout of the image was taken to see if the decrease in the peak distance was uniform, however, further analysis showed that the decrease in distance was not uniform. (See Fig - 37). Further measurement needs to be performed to see if this is an experimental error.

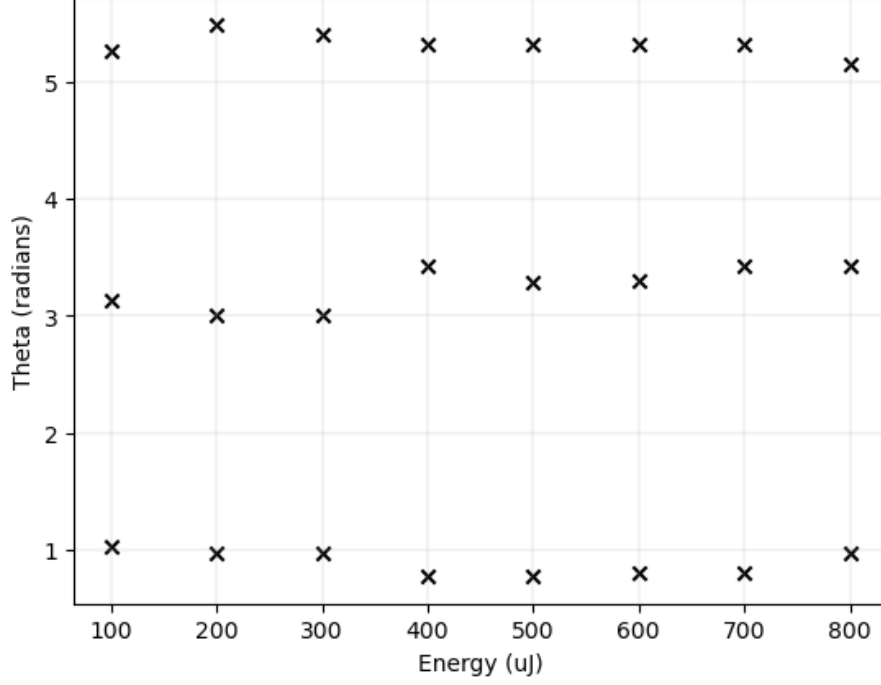


Figure 37: The peak values of the image obtained with increasing energy.

4.2.4 Polarization analysis

We finally studied the effect of circular polarization during spectral broadening. A quarter wave plate placed after the vortex can create an elliptically polarized beam. Measurements were taken while scanning from linear to circular polarization of light, to see how it affected the broadening inside the MPC. A 2D map of the spectrum as a function of the waveplate angle obtained is shown in the Fig-38

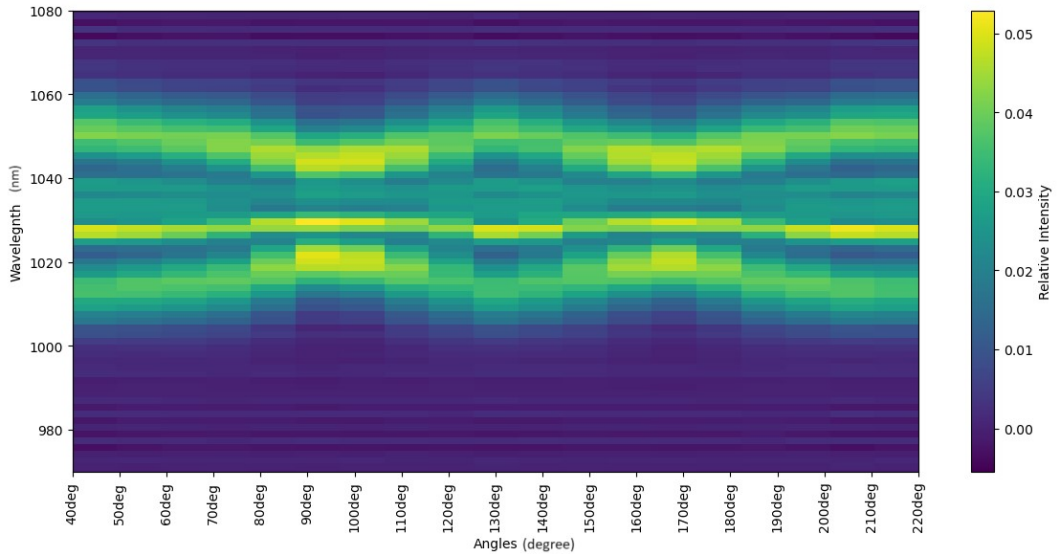


Figure 38: Intensity change in the measurement for different polarization angle.

The spectrum obtained was normalized by integrating the area under the intensity curve, in order to compare the shape of the spectrum for different angles.

The neutral axis of the waveplate is at 40° with respect to the rotational mount, meaning that for this angle the beam is linearly polarized.

We see that the broadening is stronger for regions of linear polarization, and is minimal when the incoming light is circularly polarized. The pattern repeats itself for opposite polarizations.

Finally, we made an attempt to find evidences of Spin-Orbit coupling, that is whether or not the combination of circularly polarized light and orbital angular momentum led to different broadening behavior. The difference between opposite polarization was taken, with respect to 130° which marks the perpendicular axis. The difference obtained is in Fig.39

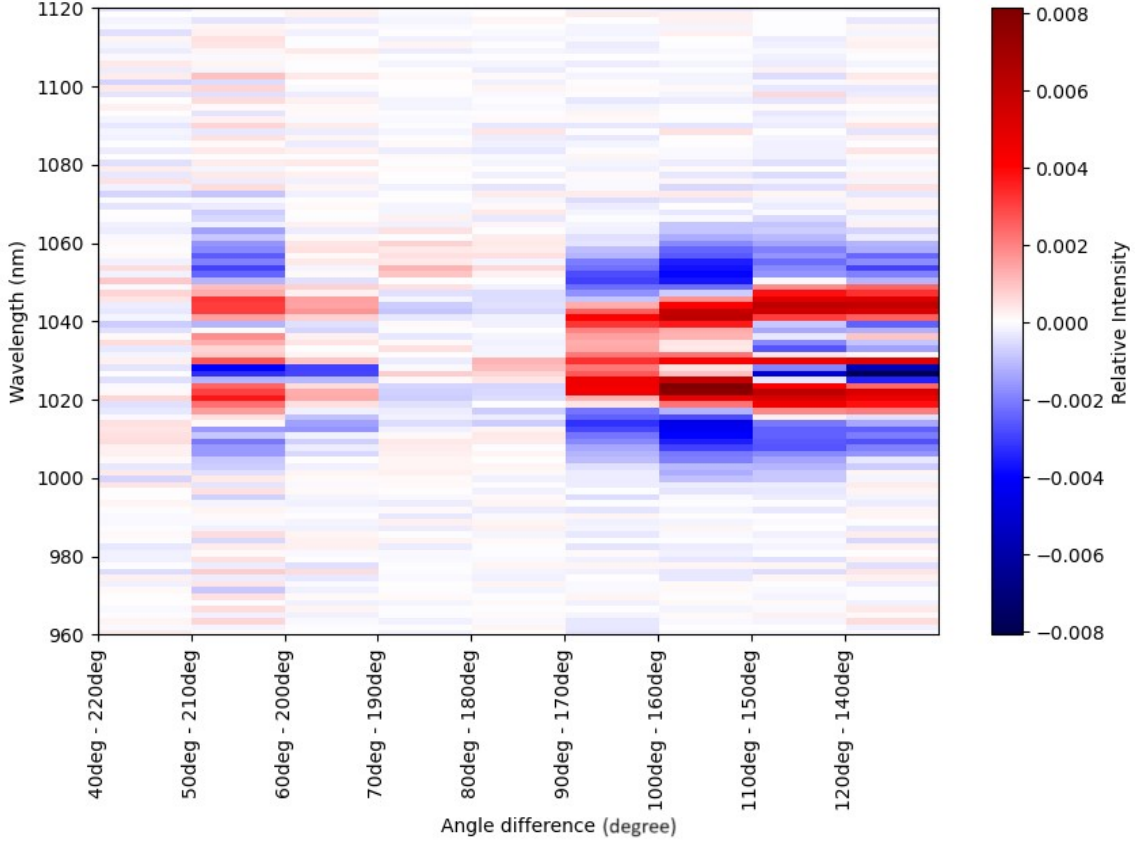


Figure 39: Difference in opposite polarization normalized with the area under the intensity curve.

The difference between two linear polarizations gives no intensity signal which is to be expected. Between the two circular polarizations (left and right), which here is around 80° we see again that the signal is zero. However, as we increase the angles to go to elliptical polarization, we see some non-zero signals arising. This can be due to two main reasons. One can be, the result of polarizing /anisotropic optics, leading to changes in the polarization. The second reason can be the vortex causes a spin-orbit coupling, leading to a broadening in the MPC. However further measurement needs to be done to cancel out the former reason before we could conclude.

5 Conclusion

This internship successfully developed a High Harmonic Generation (HHG) source, utilizing a new Ytterbium (Yb)-based femtosecond laser. The post-compression of the laser beam with a central wavelength of 1030 nm using a Multi-Pass Cell was demonstrated. The experiments were conducted in the optimal condition for an energy of 800 μ J and 2.8 bar pressure in the vacuum chamber, we achieved a pulse duration of 23.5 fs and a Fourier limit value of 21.5 fs.

We generated successfully harmonics order from $q=17$ to $q=49$. This was done for a repetition rate of 1kHz and 800 μ J energy per pulse. The spectrum obtained was in agreement with the theory of HHG.

Experiments were carried out for light carrying orbital angular momentum. The key objective behind these experiments was to find evidence of Spin-Orbit Coupling, that is with the beam carrying OAM if it leads to different broadening behavior. And we did see some variation in the spectrum obtained. We are conducting more experiments to have concrete evidence for this coupling.

These experiments are the first steps to use the full beamline with XUV pulses at high repetition rates and with controlled optical angular momentum. In the future, these will be used for spin- and angle-resolved photoemission experiments.

6 Perspectives

The beamline used during this internship is made for performing photoemission spectroscopic measurements. This spectroscopy aims to study the electronic structure of solids. Especially, the Angular Resolved Photoemission Spectroscopy, which collects information about the binding energy E_b and momentum $\hbar\mathbf{k}$ of the electron in a material by measuring kinetic energy and the angular distribution of electron photoemitted from a sample illuminated by XUV photons. The principle of ARPES is based on the photoelectric effect. The photon impinged on the material is absorbed by the electron, allowing it to escape with the maximum kinetic energy $E_{kin} = h\nu - \phi$, where ν is the photon frequency, and ϕ is the work function. Electrons emitted are collected by a spectrometer and further analyzed.

The length an electron can travel in a material without being inelastically scattered is called an inelastic mean free path. It depends on the electron kinetic energy in the material. For photon energy in the XUV range the mean free path is of the order of only a few nanometers. This is why we describe the photoemission process as a surface-sensitive process. It implies to be in the ultra-high vacuum regime where the pressure $p < 10^{-9}$ mbar.

Fig- 40 shows the schematics of the beamline in AttoLAB, for performing ARPES

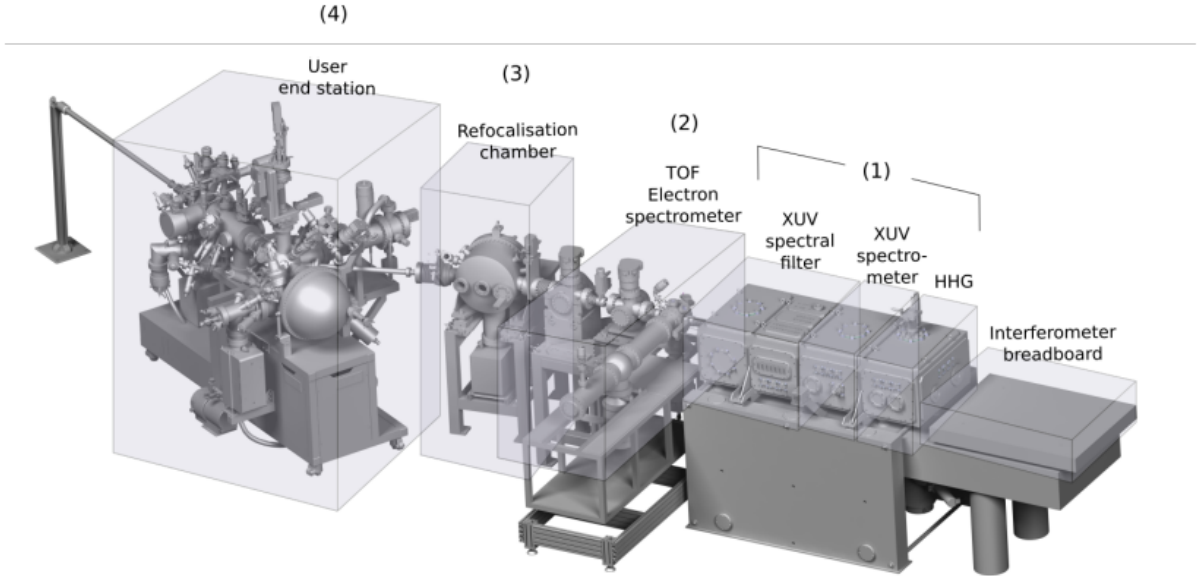


Figure 40: Beamline employed for ARPES. [47]

To spectrally manipulate the XUV HHG beam, XUV spectral filter implements there are three branches of Beamline. They are Very Broad Band (VBB), Narrow Band (NB), and Broad Band (BB). For ARPES, we employ the NB branch A.2. The branch is designed for monochromatization of the XUV with sub-harmonic spectral resolution (~ 200 meV).

Currently, our aim is to increase the XUV flux going into the Refocalisation chamber and, consequently, the user end station, to have an amplified signal. For this, we are measuring the XUV-induced photocurrent on a gold-coated toroidal mirror. We are in the process of increasing the current to around 100 pA.

For the post-compression chamber, the measurements were acquired until 3 bar pressure, and within this pressure range, we got an optimized pulse duration of $800 \mu\text{J}$ at 100 kHz. One perspective is to move to $400 \mu\text{J}$ at 200 kHz, for an even higher acquisition rate. For the energy of $400 \mu\text{J}$, implying a low energy pulse, we need to increase the pressure to get a more broadened spectrum, so as to have higher nonlinear effects.

For experiments involving light-carrying Angular Momentum, we plan to take measurements to correct the experimental errors. This involves placing a waveplate after the MPC and checking the polarization of the beam and taking measurements without the vortex plate, in order to see if the difference between polarization still exists. Another development that can be employed in the future is to have a motorized

polarizer, which can be coded using PyMoDAQ. This would ensure that no personal errors are involved while changing the polarization of the beam using the beam plate, and for easy acquisition of the spectrum after each change of angle in the polarizer. These implementations are crucial in canceling out the effects of anisotropic optics to conclude the presence of spin-orbit coupling due to nonlinear effects inside the vacuum chamber.

A Appendix

A.1 Declaration of Anti-Plagiarism

I declare that the assignment submitted is entirely my own work. The figures and information taken from other sources have been properly cited.

Krishnapriya Kozhiparambil Sajith

A.2 Experimental setup for the ARPES

The Narrow Band beamline employed for ARPES is in Fig-41

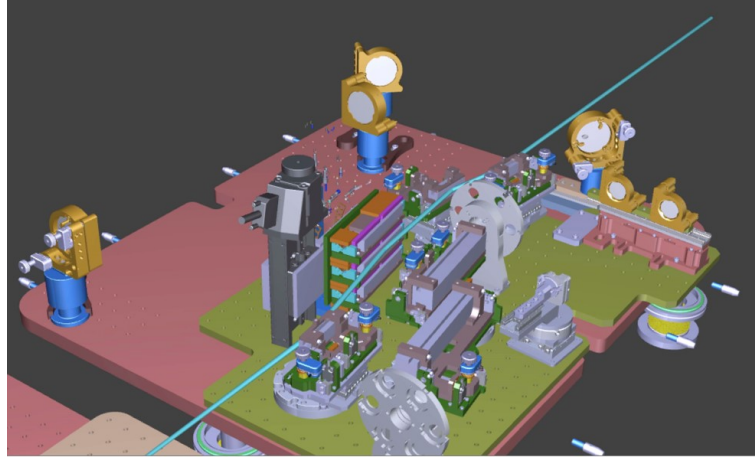


Figure 41: Narrow Band beamline setyp in AttoLAB. [47]

The Narrow Band branch has four main optical elements. The beam propagates after reflecting from a toroidal mirror (TM), then a Grating, to a plane mirror, and finally another toroidal mirror. The beam propagates into a Time Of Flight (TOF) electron spectrometer, and further into the Refocalisation chamber, where the toroidal mirror is connected to a PicoAmmeter to measure the current from photoemission. The beam further propagates to the user end station. Each of these chambers is isolated from the rest of the vacuum system.

The entire FAB10 beamline is schematized in Fig:42

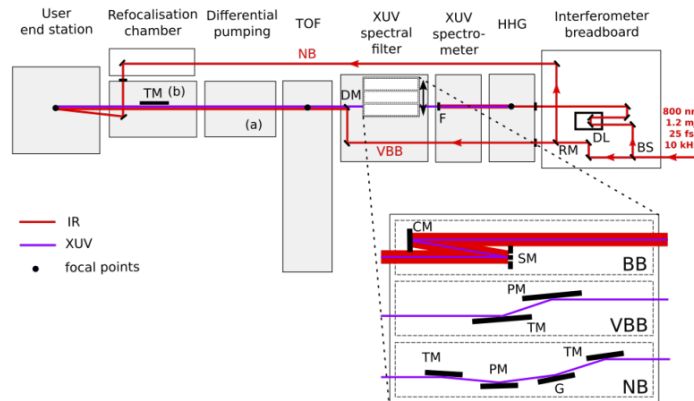


Figure 42: Optical Scheme of FAB10 - AttoLAB. The vacuum chambers are represented in grey blocks. BS: Beam Splitter. DL: delay line, RM: removable mirror (used in NB mode), F: metallic filter, DM: drilled mirror, CM: curved mirror, SM: split mirror, PM: plane mirror, TM: toroidal mirror, G: gratings[47]

The flux measurement is done by measuring photocurrent in the last toroidal mirror.

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