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high-order harmonics generation.

TESIS DOCTORAL

Agustín Carlos González Fernández

Licenciado en Ciencias Físicas
por la Universidad de Valladolid

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Ya he terminado de escribir esta tesis y estoy escribiendo su última página, la de la nostalgia, la única que incita a escribirse con el corazón, la última.

Recuerdo un día cuando empecé, hace ya un tiempo, que estaba sentado en el bar con Alberto y Cotelo ...

Resumen

A mediados de los años ochenta se observó por primera vez la aparición de los armónicos de alto orden mediante la interacción de un gas noble con un láser. Desde entonces, su importancia en la Física no ha dejado de crecer. Los armónicos de alto orden abren una ventana a los láseres de rayos X. Actualmente existen varias fuentes de rayos X que producen un elevado número de fotones como: los sincrotrones y la aceleración de electrones libres. Sin embargo aunque los láseres de sobremesa tienen un flujo menor de fotones son los que ofrecen una fuente de rayos-X de mayor calidad.

La importancia de esta fuente es múltiple. La agrupación de diversos armónicos ha permitido conseguir los pulsos electromagnéticos mas breves de la historia. Esto abre la posibilidad de estudiar los movimientos de un electrón en un átomo, así como, las velocidades de ciertas reacciones químicas. Otra de línea, es el estudio del dicroísmo en moléculas mediante el uso de láseres de rayos X polarizados circularmente.

La investigación llevada a cabo en esta tesis esta orientada hacia los láseres de sobremesa con el objetivo de llegar a mejorar su eficiencia en la generación de armónicos. Con el este fin, se desarrollaron dos códigos: uno de ellos permite conocer la interacción del láser con un gas que sale expelido por una válvula. El otro es un código que permite estudiar la interacción de un láser con un átomo. Ambos códigos estarán relacionados, ya que el primero utilizará el segundo para los cálculos atómicos necesarios cuando el láser incide sobre el gas.

Así pues, en esta tesis se presenta:

- Un capítulo introductorio que relata el desarrollo histórico de la generación de los armónicos de alto orden. Contiene un resumen de las aportaciones mas relevantes en este campo.
- Desarrollo teórico. En él se introducen los fundamentos matemáticos y físicos que sirven como base para el cálculo de los armónicos, así como las

aproximaciones aplicadas: aproximación dipolar, aproximación para un único electrón, transiciones directas ligado-libre, aproximación de campo fuerte, aproximación no relativista. Mediante la teoría de operadores de evolución temporal se llega al ecuación de Lewenstein, que es una ecuación mecano-cuántica que permite obtener: la amplitud de una transición de un electrón en un campo intenso, y el momento dipolar; el cual es la clave en el estudio de los armónicos de alto orden. Utilizando las aproximaciones anteriores, la ecuación admite una solución analítica.

Una pieza fundamental de este modelo es el cálculo de la expresión analítica de la matriz de transición dipolar. En este capítulo se muestra como obtener esta función partiendo de las diversas expresiones matemáticas que hay para la función de onda. La forma matemática de las funciones de onda de Gordon-Volkov que describen el movimiento de un electrón libre en un campo electromagnético, resultan cruciales para el cálculo analítico de la matriz de transición, ya que permite obtener una solución por medio de las transformadas de Fourier. La resolución de la integral del momento dipolar mediante la técnica de la fase estacionaria, da lugar a la aparición de las *trayectorias cuánticas*; las cuales permiten tratar todo tipo de problema de interacción entre un láser y un átomo mediante un numero limitado de trayectorias clásicas del electrón. Esta técnica es muy poderosa, ya que permite conocer cuales son los mecanismos para la producción de los armónicos y poder modificarlos.

- Mediante la mezcla de dos gases con diferente energía de ionización como el helio y el neón, y utilizando la técnica de las trayectorias cuánticas, se analiza cual tiene que ser el ratio de esta mezcla para modificar la intensidad de un armónico. Este estudio ha precisado desarrollar una nueva matriz de transición dipolar, puesto que la utilizada en la bibliografía genera una fase errónea para el dipolo. Los resultados obtenidos se han contrastado con datos experimentales hasta donde ha sido posible. Para las regiones del espectro donde no había datos experimentales, se ha comparado con resultados obtenidos con un solver TDSE. Como resultado, se ha publicado un artículo en una revista científica de primer orden.
- El estudio del dicroísmo se realiza mediante láseres de rayos X con polarización circular. Para este fin, se ha desarrollado una nueva matriz de transición dipolar para el neón, que incluye los números cuánticos mag-

néticos. Este estudio ha proporcionado algunos resultados sorprendentes, en un láser con polarización ortogonal ω , 2ω los dos electrones con números cuánticos $m = \pm 1$ producen armónicos pares e impares, pero debido a la interferencia destructiva algunos son anulados. Con este modelo se ha estudiado como afecta la forma de la envolvente del campo eléctrico a la elipticidad de los armónicos, demostrando que sólo los campos con una envolvente no constante son capaces de producir armónicos circulares.

- Finalmente, se aborda la interacción del láser con un gas. En este capítulo se establecen los fundamentos teóricos para la propagación de un campo electromagnético en un medio gaseoso. Se ha desarrollado una expresión matemática para el cálculo de la susceptibilidad para el caso de láseres de alta intensidad. Las simulaciones computacionales muestran como se modifica la propagación del láser en un gas cuando se ioniza. También como depende la emisión de los armónicos modificando algunos parámetros como, la presión del gas, la intensidad del láser, la posición del foco. La comparación con resultados experimentales ofrece un buen grado de concordancia.

Abstract

In the mid-1980s, for the first time, the appearance of high order harmonics was observed through the interaction of a noble gas with a laser. Since then, its importance in physics has not stopped growing. High-order harmonics open a window to X-ray lasers. Currently there are several sources of X-rays that produce a high number of photons, such as synchrotrons and acceleration of free electrons. However, although desktop lasers have a smaller flow of photons, they offer a higher quality X-ray source.

The importance of this source is manifold. The grouping of different harmonics has made it possible to get the shortest electromagnetic pulses in history. This opens the possibility of studying the movements of an electron in an atom, as well as the speeds of certain chemical reactions. Another line, is the study of dichroism in molecules through the use of circularly polarized X-ray lasers.

The research carried out in this thesis is oriented towards the desktop lasers with the aim of improving their efficiency in the generation of harmonics. With this purpose, two codes were developed: one of them allows to know the interaction of the laser with a gas jet. The other is a code that studies the interaction of a laser with an atom. Both codes will be related, since the first one will use the second one for the necessary atomic calculations when the laser interacts with the gas.

Thus, in this thesis it is presented:

- An introductory chapter that tells the historical development of the generation of harmonics of high order. It contains a summary of the most relevant contributions in this field.
- Theoretical development. It introduces the mathematical and physical fundamentals that serve as the basis for the calculation of harmonics, as well as the approximations applied: dipolar approximation, single atom electron approximation, direct bound-free transitions, strong field approx-

imation, non-relativistic approximation. By means of the theory of the time evolution operator we arrive at the Lewenstein equation, which is a quantum equation that allows to obtain: the amplitude of a transition of an electron in an intense field and the dipole moment; which is the key in the study of high order harmonics. Using the above approximations, an analytical solution of the equation is possible.

A fundamental part of this model is the calculation of the analytical expression of the dipolar transition matrix. This chapter shows how to derive this function from the various mathematical expressions for the wavefunction. The mathematical form of the Gordon-Volkov wave functions that describe the movement of a free electron in an electromagnetic field, are crucial for the analytical calculation of the transition matrix, since it allows to obtain a solution by means of the Fourier transform. The resolution of the integral of the dipole moment by means of the technique of the stationary phase, gives rise to the appearance of the *quantum trajectories*; which allow to treat all type of problem of interaction between a laser with an atom by means of a limited number of classic trajectories of the electron. This technique is very powerful, since it allows to know which are the mechanisms for the production of harmonics and to be able to modify them.

- By mixing two gases with different ionization energy such as helium and neon, and using the technique of quantum paths, we analyze what must be the ratio of this mixture to modify the intensity of a harmonic. This study has needed to develop a new dipolar transition matrix, since the one used in the literature generates an erroneous phase for the dipole. The results obtained have been compared with experimental data to where possible. For regions of the spectrum where no experimental data were available, it has been compared with results obtained with a TDSE solver. As a result, an article has been published in a leading scientific journal.
- The study of dichroism is done by X-ray lasers with circular polarization. To this end, a new dipole transition matrix for neon has been developed, including magnetic quantum numbers. This study has provided some surprising results, for a laser with orthogonal polarization ω 2ω , the two electrons with quantum numbers $m = \pm 1$ produce even and odd harmonics, but due to destructive interference some are annihilated. With this

model has been studied how it affects the shape of the envelope of the electric field to the ellipticity of harmonics, showing that only fields with a non-constant envelope are capable of producing circular harmonics.

- Finally, the interaction of the laser with a gas is tackled. This chapter establishes the theoretical foundations for the propagation of an electromagnetic field in a gaseous medium. A mathematical expression has been developed for the calculation of susceptibility in the case of high intensity lasers. Computational simulations show how the propagation of the laser in a gas is modified when it is ionized. Also as it depends the emission of the harmonics modifying some parameters like, the pressure of the gas, the intensity of the laser, the position of the focus. The comparison with experimental results offers a good degree of agreement.

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Chapter 1

Introduction

When an electric field with enough intensity interacts with an atom, some times there are involved processes with more than a single photon. This multiphoton processes can be classified as: multiphoton excitation, collisional-radiative in presence of a laser, multiphoton ionization (MPI) where a atom absorbs several photons and one or more electrons are ionized, and high harmonics generation where several photons are absorbed and a single photon is emitted with a frequency of the harmonic of the laser.

For hydrogen atom, the field strength E_a is given by

$$E_a = \frac{e}{(4\pi\epsilon_0)a_0^2} \approx 5.14221 \times 10^{11} \text{V/m} \quad (1.1)$$

where e is the absolute electron charge, a_0 is the Bohr radius. The intensity of a laser that corresponds to above field strength is $I_a = \epsilon_0 c E_a / 2 = 3.50945 \times 10^{16} \text{W/cm}^2$, therefore, for lasers with intensities $I \ll I_a$, the interaction atom-laser can be studied by a perturbative theory, where the interaction term in the Schrödinger equation is treated as a perturbation. When the intensity of the laser is of the order of I_a or exceeding, it must be analyzed as a non-perturbative approaches.

The MPI is represented by the reaction $n\hbar\omega + A^q \rightarrow A^{q+1} + e$ when an electron is ionized by means of absorption of n photons. This processes was observed in the 1960's where the laser intensity was $I \ll I_a$. In the experiment was observed that the total n-photons of the ionization rate were proportional to I^n . Experiments at later of 1970's with higher intensities $I > 10^{11} \text{W/cm}^2$ shown that the freed electron can absorb more photons that the minimum number required for the ionization. This processes were called above threshold ionization (ATI). The spectra of photo-electrons consists in several peaks separated by $\hbar\omega$

Figure 1.1: Figure (a): multiphoton Ionization (MPI): The electrons absorbs several photons in intermediate virtual states until reach the ionization energy. I_p Figure (b): Above threshold Ionization (ATI): The electrons absorbs more photons than in MPI and is free with a kinetic energy given by the extra photons.

laser frequency

$$E_s = (n_0 + s)\hbar\omega - I_p \quad (1.2)$$

where n_0 is the minimum photon required for ionization, s is the excesses of photons absorbed by the electron, I_p ionization energy. One of the consequences of the ATI processes was the ionization rate is not proportional to I^{n_0+s} .

The freed electrons are under interaction of the electric field with an oscillation movement that follows the electric field. The cycle-average of the kinetic energy is given by

$$U_p = \frac{e^2 E_0^2}{4m_e \omega} \propto \frac{I}{\omega^2} \quad (1.3)$$

where U_p is the kinetic energy and usually is named as ponderomotriz energy, E_0 is the electric field strength, m_e electron mass, ω electric field frequency.

When the frequency of the electric field is low enough and the intensity is sufficiently strong, the ionization will be able interpreted by using a quasi-static model in which the bound electrons experience an effective potential formed by adding the atomic potential to the contribution of the external electric field. The ionization rate for hydrogenic systems may be approximated by the static-limit tunneling rate calculated by Ammosov, Delone and Krainov (ADK) [3]. For hydrogenic system in the ground state, the tunneling rate is given in atomic units.

$$W_{ion} = \frac{4(2I_p)^{5/2}}{E} \exp\left(-\frac{2(2I_p)^{3/2}}{2E}\right) \quad (1.4)$$

Figure 1.2: Figure (a): When the electric field is zero, the electron is confined to the atomic potential $V(x)$ with a ionization energy of I_p . Figure (b): When the electric field increases the electrons through the potential barrier, $I \ll I_a$. Figure (c): When the intensity of the electric field is greater than the critical intensity $I \gg I_c$, the electron is free, barrier suppression (OBI)

where E is the static electric field strength. Above expression indicates that the tunneling occurs predominantly at peaks of electric field, when the superposition of the potential barrier and electric field has its lower value. Therefore, the electrons are emitted as burst over time.

In the 1960s, Keldysh[55] developed a quasi-static theory to describe the multiphoton ionization. The theory describes the photo-ionization by strong laser when the photon energy of the laser is much smaller than the ionization energy. He derived a photo-ionization expression for the transition ground-free state, where the free state was represented by the Volkov wave-functions, which represent the electron movement in a electric field by wave-planes. In this first theory, no states were taken between the free and bound states, and the resonance states as well as the interaction of the Coulomb potential in the continuous states were neglected.

This approximation was known as the Strong Field Approximation (SFA). An important quantity in this theory is the Keldysh parameter γ_k , defined as the ratio of the laser and tunneling frequencies, which is given by

$$\gamma_k = \left(\frac{I_p}{2U_p} \right)^{1/2} \quad (1.5)$$

For $\gamma_k \lesssim 1$, tunneling dynamics dominates, while $\gamma_k \gtrsim 1$ is referred to the multiphoton ionization regime.

At low frequencies, when the laser intensity increases the effective potential barrier is lower and narrow, and eventually, when the laser reaches the critical

intensity I_c , the electron can flow freely from the atom to the continuum. This is known as the over the barrier ionization (OBI). The critical intensity I_c for the Hydrogen is $1.4 \times 10^{14} \text{Wcm}^{-2}$ and for the Helium $1.5 \times 10^{15} \text{Wcm}^{-2}$.

Figure 1.3: The three step model. First step the ionization at time τ_i , latter on propagation as free particle in the electric field, finally recombination at time τ_r with the emission of a photon with energy $\hbar\omega$

A model for strong field at low frequencies needs to take into account the possibility that a ionized electron can return to the parent ion. A semi-classical model developed by Corkum [31] and by Kulander, Schafer and Krause [60] is based on the idea that the ionization has several steps. In the first step, the electron in ground state is separated from its parent ion by tunneling or the over barrier ionization processes. The second step, the electron is free and interacts with the electric field by means of classical mechanism. If the electron does not return to the parent ion, then occurs the ionization. On the contrary, when it returns, the third step, the recombination leads the emission of the high-harmonics. This model was named as *the three step model*.

When the atoms interact with sufficiently intense laser, they emit radiation which frequency is a harmonic of the fundamental frequency of the laser. For lasers linearly polarized with a single central frequency, the harmonics emitted by the atoms are an odd number of the laser frequency. When the laser has a few cycles, the spectrum is a continuous distribution of frequencies, not a set of discrete frequencies such as will happen for long pulses. On the other hand, when the laser has large pulses, it provokes the ionization of the matter, reducing the efficiency of the harmonics generated. The high-order harmonics generation (HHG) provides a source at high-frequency, coherent and bright.

In the late of 1980's the availability of lasers with sufficiently intensity and

Figure 1.4: Typical spectrum, the plateau is extended from the lowest frequency that corresponds to I_p to the harmonic ω_c . In the cut-off regions, the intensity of the harmonics beyond ω_c decrease rapidly.

short pulses made the possibility to observe the HHG. Several experiments in the first half of 1990's allowed to observe high-order harmonics using different noble gases. The order of harmonics was increased with each new experiment, using a wide range of length pulses and intensities of the lasers. The experiments shown a region of the spectrum with constant conversion efficiency, known as plateau. The existence of the plateau only could be explained with non-perturbative theories. The increase of intensity permits the generation of photons with much higher frequencies. Chang et al. [28] with a laser of Ti:sapphire centered at $\lambda = 800 \text{ nm}$, with 26 fs pulse duration, reached harmonics with wavelengths down to $\lambda = 2.7 \text{ nm}$ (460 eV) for the Helium, and $\lambda = 5.2 \text{ nm}$ (239 eV) for the Neon. For the Helium, discrete harmonic peaks was observed up to the 221^{th} order. These wavelengths are within the “water window” region of X-ray transmission. Solving numerically the Time Dependent Schrödinger Equation (TDSE), Krause, Schafer and Kulander [59] found that there is a frequency ω_c in the harmonic spectrum which indicates the highest harmonic from the plateau. The harmonics beyond ω_c rapidly decrease their intensity. Its value is given by

$$\omega_c \approx (I_p + 3U_p)/\hbar \quad (1.6)$$

Using a semi-classical model, Corkum [31] calculates the maximum energy possible of the harmonic was $I_p + 3.17U_p$. Latterly this was confirmed by new calculation and experiments.

Figure 1.5: Recombination and ionization times from short and long trajectories, compared to the temporal evolution of the electric field. The abscissa axis corresponds to the emitted harmonic at the recombination time.

Based on the SFA, Lewenstein et al. [64, 1] developed a quantum mechanical model for HHG, which permits a faster calculations compared to the TDSE, and also provides a deeper insight in the process of harmonics generation. Balcou [10, 8] described that the phase ϕ of the dipole moment was proportional to τU_p , where τ is the time spent by electron between the ionization and the recombination. According to Eq. (1.3), the initial phase at the recombination time is proportional to the laser intensity $\phi \propto I$. The emission of each harmonic from the plateau has the contribution of two electron trajectories, referred as short and long trajectories that describe the time spent by the electron from the ionization to recombination. On the other hand, beyond the cut-off only a single trajectory contributes the the spectrum.

The study of the harmonic generation by an intense laser on a gaseous medium is divided in two parts. The first covers the response from a single-atom to the laser, over a wide range of intensities. The second aspect is related to the solution of the Maxwell equations in the gaseous medium for the electric fields of the laser and the harmonics. The interference between the harmonics generated along the medium can attenuate a particular harmonic. The modification of the phase-matching conditions is crucial to improve the efficiency of the harmonics.

Chapter 2

Theoretical Model.

Although the high-intensity laser-atom interaction can be tackled by solving numerically the time-dependent Schrödinger equation (TDSE), the time that it takes to calculate the spectrum of a single atom is rather long. It becomes a problem when TDSE is combined with the calculation of several spectra to solve numerically the phase-matching problem that appears in the interaction with a jet of gas, such as is reported in the chapter 5.

Lewenstein and his collaborators demonstrated [64, 1] that the equation can be solved analytically under the strong field approximation (SFA). The tool is not only fast, also provides a clear explanation of the physical mechanisms of the interference processes between the short and long trajectories that finally produces the typical spectrum.

A section of the chapter is dedicated to obtaining the expression known as the Lewenstein equation. The procedure is different from the one originally followed in the paper [64], and is based on time-evolution operators [19].

The chapter is organized as follows. The first section is about how the gauge transformations of the electric field modifies the shape of the wavefunction. Although there is a great variety of gauge transformations, here is only treated the three of them: the length, velocity, and Kramers-Henneberger gauges. Bauer, Milošević, and Becker demonstrate that in the SFA only the length gauge coincides with the exact numerical solution of TDSE, meanwhile the velocity gauge of the ground states with odd parity differs with the length gauge qualitatively. [15]. On the other hand, the velocity gauge is preferred to obtain the solutions of TDSE, section 2.5.5

We can see that the mathematical expressions of the interaction term of the Hamiltonian are different depending on the selected gauge.

In the following section is dedicated to the Gordon-Volkov wavefunctions. This is a key component of the Lewenstein model because when the electron is freed only the external field interacts with it. The field generated by the parent ion is neglected, regardless the distance between the electron and the parent ion. Other studies [93] use continuum states modified by the coulomb potential instead the Gordon-Volkov wavefunction, to include the Coulomb interaction within the wavefunction. After an extensive development of the time-evolution operator, we reach the section where the dipole moment is calculated, that is analytically solved by means of several approximations. First, we develop an analytic expression of the dipolar transition matrix. Subsequently, by means of the saddle point, the integral of the dipole moment is transformed in a summation in which each term corresponds to a short or long trajectory of the electron in the continuum.

The saddle point approximation has been widely used in the literature with a great success in the interpretation of experimental results like the influence of the carrier-envelope phase in the production of HHG of Sansone et al.[87], or the description of the contribution to the HHG from the continuum-continuum transitions of Kohler et al. [57].

A small section is dedicated to the TDSE (Qprop) where the fundamentals of this code are presented schematically. The results of Qprop is used intensively in the following chapters in most cases as a reference.

The last part treats with several tools used to analysis the results: the time-frequency analysis (TFA) based on the Gabor transformation, and the inverse Fourier transformation to see how is the temporal evolution of pulse formed by the contribution of a range of harmonics.

2.1 Dipole approximation.

This approximation is based on the wavelength of the laser is large compared with the size of the atom, and the field strength is not too high. When both conditions are covered it is possible to neglect the spatial variation of the electric field through the atom.

In this approximation, for an atom whose nucleus is located at the position $\mathbf{r}$, the vector potential $\mathbf{A}(\mathbf{r}, t) = \mathbf{A}(t)$ is spatially homogeneous, is to say, it depends only on the time variable, so that the Coulomb gauge condition is automatically satisfied $\nabla \cdot \mathbf{A}(t) = 0$.

2.2 Gauge transformations.

The resolution of the Schrödinger equation is often complex, but we can rely on some approximations and transformations to make this task simpler. The main difficulty of the transformations is to generate valid wavefunctions, that is, whose measurable magnitudes are not altered by the transformation. Therefore, the transformation of the wavefunction must be preserves the norm of the function but not necessarily its phase. On the other hand, the electric and magnetic field are not univocally determined by their potentials.

Then, the type of transformation must fulfill the invariance of the Maxwell equations and the norm of the wavefunction.

The interaction Hamiltonian.

From now onward, all equations are in atomic units $m_e = -e = \hbar = 1$. The interaction Hamiltonian of an electron in an electromagnetic field in its more general form is given by

$$\begin{aligned} H(t) &= \frac{1}{2} (\mathbf{p} + \mathbf{A})^2 - V \\ &= \frac{\mathbf{p}^2}{2} + \frac{1}{2} (\mathbf{A} \cdot \mathbf{p} + \mathbf{p} \cdot \mathbf{A}) + \frac{1}{2} \mathbf{A}^2 - V \end{aligned} \quad (2.1)$$

where $V(\mathbf{r}, t)$ is the scalar potential, and $\mathbf{A}(\mathbf{r}, t)$ is the vector potential. So, the spinless Schrödinger equation for an electron is given by

$$i \frac{\partial}{\partial t} \Psi(\mathbf{r}, t) = \left[-\frac{1}{2} \nabla^2 - \frac{i}{2} (\mathbf{A} \cdot \nabla + \nabla \cdot \mathbf{A}) + \frac{1}{2} \mathbf{A}^2 - V \right] \Psi(\mathbf{r}, t) \quad (2.2)$$

where $\mathbf{p} = -i\nabla$. And using the following relations

$$\begin{aligned} \nabla \cdot (\mathbf{A}\Psi) &= \mathbf{A} \cdot (\nabla\Psi) + (\nabla \cdot \mathbf{A})\Psi \\ &= \mathbf{A} \cdot (\nabla\Psi) \end{aligned} \quad (2.3)$$

where last equality uses the fact that $\nabla \cdot \mathbf{A} = 0$. That is implicitly covered by the dipole approximation. Finally, we get

$$i \frac{\partial}{\partial t} \Psi(\mathbf{r}, t) = \left[-\frac{1}{2} \nabla^2 - \frac{i}{2} (\mathbf{A} \cdot \nabla) + \frac{1}{2} \mathbf{A}^2 - V \right] \Psi(\mathbf{r}, t) \quad (2.4)$$

Gauge transformation.

The invariance of $\mathbf{E}$ and $\mathbf{B}$ under the gauge transformation in the Maxwell equations allows imposing various conditions on the potentials V and $\mathbf{A}$. The selection of the Coulomb gauge assumes $\nabla \cdot \mathbf{A} = 0$ and $V = 0$ for the empty space.

The TDSE in Eq. (2.2) in presence of the scalar and vector potentials $V(\mathbf{r}, t)$ $\mathbf{A}(\mathbf{r}, t)$ can be maintained unchanged under the following gauge transformation

$$\begin{aligned}\mathbf{A} &\rightarrow \mathbf{A}' = \mathbf{A} + \nabla f \\ V &\rightarrow V' = V - \frac{\partial f}{\partial t}\end{aligned}\tag{2.5}$$

$$\Psi(\mathbf{r}, t) \rightarrow \Psi'(\mathbf{r}, t) = \Psi(\mathbf{r}, t) \exp(-if)$$

here, f is an arbitrary differentiable function of $\mathbf{r}$ and t . Then, the wavefunction Ψ' satisfies the equation

$$i \frac{\partial}{\partial t} \Psi'(\mathbf{r}, t) = \left[-\frac{1}{2} \nabla^2 - \frac{i}{2} (\mathbf{A}' \cdot \nabla + \nabla \cdot \mathbf{A}') + \frac{1}{2} \mathbf{A}'^2 - V' \right] \Psi'(\mathbf{r}, t) \tag{2.6}$$

where both equations (2.2) and (2.6) have the same form, and the wavefunction only changes by a complex correction factor. So, a suitable election of the gauge in Eq. 2.2 can simplify the interaction term of the equation, and then, the wavefunction only changes the phase. It is often convenient to work in the Coulomb gauge.

Velocity gauge.

The term $\mathbf{A}^2$ can be removed in Eq. (2.6) extracting its corresponding phase from the wavefunction by the unitary transformation on the wavefunction given by

$$\Psi^V(\mathbf{r}, t) = \exp \left[\frac{i}{2} \int_{-\infty}^t \mathbf{A}^2(t') dt' \right] \Psi(\mathbf{r}, t) \tag{2.7}$$

where the function f should be the following form

$$f = -\frac{1}{2} \int_{-\infty}^t \mathbf{A}^2(t') dt' \tag{2.8}$$

with this choice, the potentials are given done by

$$\begin{aligned}\mathbf{A}' &= \mathbf{A} \\ V' &= V + \frac{1}{2} \mathbf{A}^2\end{aligned}\tag{2.9}$$

The new TDSE with the new wavefunction including change $\Psi \rightarrow \Psi^V$ is the following

$$i \frac{\partial}{\partial t} \Psi^V(\mathbf{r}, t) = [H_0 + \mathbf{A}(t) \cdot \mathbf{p}] \Psi^V(\mathbf{r}, t) \quad (2.10)$$

where H_0 is Hamiltonian operator in the ground state.

$$H_0 = \frac{\mathbf{p}^2}{2} + V \quad (2.11)$$

Eq. 2.10 is known as velocity gauge, since the interaction Hamiltonian is given by

$$H_{int}^V(t) = \mathbf{A}(t) \cdot \mathbf{p} \quad (2.12)$$

where this Hamiltonian couples the vector potential to the momentum operator.

Length gauge.

Other form of TDSE can be obtained by performing the following gauge transformation

$$f = -\mathbf{A}(t) \cdot \mathbf{r} \quad (2.13)$$

from Eq. 2.5

$$\begin{aligned} \mathbf{A}' &= 0 \\ V' &= V + \frac{\partial \mathbf{A}}{\partial t} \cdot \mathbf{r} = V - \mathbf{E}(t) \cdot \mathbf{r} \end{aligned} \quad (2.14)$$

then, the wavefunction is

$$\Psi^L(\mathbf{r}, t) = \exp[i\mathbf{A}(t) \cdot \mathbf{r}] \Psi(\mathbf{r}, t) \quad (2.15)$$

that satisfies the TDSE

$$i \frac{\partial}{\partial t} \Psi^L(\mathbf{r}, t) = [H_0 + \mathbf{E}(t) \cdot \mathbf{r}] \Psi^L(\mathbf{r}, t) \quad (2.16)$$

above equation is denoted as the length gauge, and the interaction Hamiltonian is

$$H_{int}^L(t) = \mathbf{E}(t) \cdot \mathbf{r} \quad (2.17)$$

Kramers-Henneberger transformation.

Also known as the acceleration gauge, because it studies the interaction of an atomic system with a laser field in an accelerated frame. The wavefunction in this new frame is

$$\Psi^A(\mathbf{r}, t) = \exp[i\boldsymbol{\alpha}(t) \cdot \mathbf{p}] \Psi^V(\mathbf{r}, t) \quad (2.18)$$

where $\boldsymbol{\alpha}(t)$ is defined as

$$\boldsymbol{\alpha}(t) = \int_{-\infty}^t \mathbf{A}(t') dt' \quad (2.19)$$

where the new TDSE is given by

$$i \frac{\partial}{\partial t} \Psi^A(\mathbf{r}, t) = \left[\frac{\mathbf{p}^2}{2} + V(\mathbf{r} + \boldsymbol{\alpha}(t)) \right] \Psi^A(\mathbf{r}, t) \quad (2.20)$$

where the interaction of the laser field has been included inside the atomic potential by means $\boldsymbol{\alpha}(t)$, then V is now time dependent. Then, the central force of atomic nucleus, that is fixed in laboratory frame, now oscillates as $\boldsymbol{\alpha}(t)$.

This gauge is not very used in literature, although it offers some interesting properties. When the interaction laser-wavefunction is numerically calculated by a TDSE solver, if the intensity of the laser is high enough, the tail of the wavefunction can be pushed to the edge of the grid. Therefore, the size of the grid has to be increased. Using the KH transformation, this frame oscillates with the laser and the wavefunction follows this oscillation; it is possible to reduce the size of the grid. [81].

Another interesting application was proposed by D. Bauer [17], who used this gauge transformation to calculate the wavefunction that includes the atomic potential as a shift of the phase in the Volkov function.

Finally, Smirnova and Ivanov did some ionization studies based on this transformation [92] [48].

2.3 The Gordon-Volkov wavefunction.

Let us consider the case of a free electron in the presence of an electric field. Then, in Eq. (2.2) the atomic potential is removed. The wavefunction of the

electron with the dipole approximation for an spinless particle is governed by the Schrödinger equation

$$\begin{aligned} i\frac{\partial}{\partial t}\chi(\mathbf{r}, t) &= H_F(t)\chi(\mathbf{r}, t) \\ &= \frac{1}{2}[\mathbf{p} + \mathbf{A}(t)]^2\chi(\mathbf{r}, t) \end{aligned} \quad (2.21)$$

where $H_F(\mathbf{r}, t)$ alludes to the Hamiltonian of the free particle in the electric field. Using the velocity gauge transformation in Eq. 2.7, the new wavefunction is given by

$$\chi^V(\mathbf{r}, t) = \exp\left[\frac{i}{2}\int_{-\infty}^t \mathbf{A}^2(t')dt'\right]\chi(\mathbf{r}, t) \quad (2.22)$$

and the TDSE for previous wavefunction in the velocity gauge comes

$$\begin{aligned} i\frac{\partial}{\partial t}\chi^V(\mathbf{r}, t) &= H_F^V\chi^V(\mathbf{r}, t) \\ &= \left[\frac{\mathbf{p}^2}{2} + \mathbf{A}(t) \cdot \mathbf{p}\right]\chi^V(\mathbf{r}, t) \end{aligned} \quad (2.23)$$

The operator $\mathbf{p}$ has the eigenfunction $\exp(-i\mathbf{k} \cdot \mathbf{r})$ with the eigenvalue $\mathbf{k}$. How H_F commutes with the operator $\mathbf{p}$ the wavefunctions of the electron should have the form

$$\chi_{\mathbf{k}}^V(\mathbf{r}, t) = (2\pi)^{-3/2}\exp(i\mathbf{k} \cdot \mathbf{r})f_{\mathbf{k}}(t) \quad (2.24)$$

replacing the equation (2.24) in (2.23) the final solution is the Gordon-Volkov of the normalized wavefunction [51]

$$\chi_{\mathbf{k}}^V(\mathbf{r}, t) = (2\pi)^{-3/2}k^{1/2}\exp\{i\mathbf{k} \cdot [\mathbf{r} - \boldsymbol{\alpha}(t)] - iE_k t\} \quad (2.25)$$

where $E_k = k^2/2$ is the kinetic energy of an electron with momentum $\mathbf{k}$, and $\boldsymbol{\alpha}(t)$ see (2.19)

The Gordon-Volkov wavefunctions in the velocity gauges satisfies the following orthogonal condition

$$\langle \chi_{\mathbf{k}'}^V | \chi_{\mathbf{k}}^V \rangle = \delta(\mathbf{k} - \mathbf{k}') \quad (2.26)$$

To obtain the Gordon-Volkov equation in the length gauge, we must first obtain the untransformed original $\chi_{\mathbf{k}}(\mathbf{r}, t)$ by means of Eq. (2.22)

$$\chi_{\mathbf{k}}(\mathbf{r}, t) = (2\pi)^{-3/2}k^{1/2}\exp\{i\mathbf{k} \cdot [\mathbf{r} - \boldsymbol{\alpha}(t)] - iE_k t - \frac{i}{2}\int_{-\infty}^t \mathbf{A}^2(t')dt'\} \quad (2.27)$$

from previous equation, in the length gauge using the transformation (2.15) is

$$\begin{aligned}
 \chi_{\mathbf{k}}^L(\mathbf{r}, t) &= (2\pi)^{-3/2} k^{1/2} \\
 &\times \exp\{i\mathbf{A}(t) \cdot \mathbf{r} + i\mathbf{k} \cdot [\mathbf{r} - \boldsymbol{\alpha}(t)] - iE_k t - \frac{i}{2} \int_{-\infty}^t \mathbf{A}^2(t') dt'\} \\
 &= (2\pi)^{-3/2} k^{1/2} \\
 &\times \exp\{i[\mathbf{k} + \mathbf{A}(t)] \cdot \mathbf{r} - \frac{i}{2} \int_{-\infty}^t [\mathbf{k} + \mathbf{A}]^2(t') dt'\}
 \end{aligned} \tag{2.28}$$

where $\chi_{\mathbf{k}}^L(\mathbf{r}, t)$ is an eigenfunction of the TDSE:

$$\begin{aligned}
 i \frac{\partial}{\partial t} \chi_{\mathbf{k}}^L(\mathbf{r}, t) &= H_F^L \chi_{\mathbf{k}}^L(\mathbf{r}, t) \\
 &= \left[\frac{\mathbf{p}^2}{2} + \mathbf{E}(t) \cdot \mathbf{r} \right] \chi_{\mathbf{k}}^L(\mathbf{r}, t)
 \end{aligned} \tag{2.29}$$

and it satisfies the orthogonal condition

$$\langle \chi_{\mathbf{k}'}^L | \chi_{\mathbf{k}}^L \rangle = \delta(\mathbf{k} - \mathbf{k}') \tag{2.30}$$

2.4 The Lewenstein model.

In the early 1990s, Lewenstein developed a model that allowed to obtain the dipole moment of an electron in a simple way and from here the spectrum of high-order harmonics. This model was quickly adopted by the scientific community, since it allowed to obtain the calculations of the spectra at much higher speeds than the TDSE model.

This was decisive in the calculations of the interactions of the laser with a jet of gas [80], where the electric field changes from a region to other in the gas as a result of the laser focusing. Where to have smooth variations of the dipole moment along the gas regions, the size of the cell used in calculations should be small, and as a result, the number of calculations increase drastically. The great improvement in the calculation speed comes from the fact that the model uses an analytical expression of the transition matrix, whereas to do the same, the TDSE must calculate the integral for each of the points in the grid where the wavefunction is defined.

Perhaps, more important than the improvement in the speed of the calculation, is the possibility to see in detail some physical processes that the model of TDSE does not do. The model allowed us to treat the electron motions as if

they were classical trajectories [85], and get the dipole moment as summation of all possible trajectories that the electron could have under the interaction of the external field. This way of treating macroscopic results looks like the mathematical model developed by Feynman for quantum electrodynamics by means of *the quantum path integrals*.

First analysis shows the photon emitted by an atom is the result of the superposition of two photons with same frequency emitted at different times.

These two photons are emitted when the electron recombines with the parent ion after having followed two different trajectories as free particles under the electric field interaction. These trajectories are named as *quantum paths* or *quantum trajectories*. The quantum paths are classified as short or long depending on the difference of time between recombination and ionization.

The most important contribution to the dipole moment comes from those quantum trajectories in which the electron recombines in the first visit to parent ion. Although, the electrons can recombine in later visits. There is not significant difference from the dipole modulus point of view when the process in which the electrons recombine is in a later visit to the third visit the parent ion. The inclusion of other processes allows to know the contribution of continuous-continuous transitions in the presence of an electric field, or elastic collisions of the electrons with the atom.

Although the Lewenstein model offers wide possibilities it has several weaknesses, only dealing with the bound-free transitions, and the atomic potential is neglected even if the released electron is near the parent ion.

2.4.1 The time-evolution operator.

The original model proposed by Lewenstein is the solution of the Schrödinger equation formed by the temporal evolution of two states: the ground $|\Psi_0\rangle$ and the continuum $|k\rangle$. The ground state is multiplied by a factor $a(t)$ that corresponds to the amplitude of the ground state. The continuum states are multiplied by $b(\mathbf{k}, t)$, the density of the states in the continuum.

$$|\Psi(\mathbf{r}, t)\rangle = e^{-E_0 t i} \left[a(t) |\Psi_0\rangle + \int b(\mathbf{k}, t) |\mathbf{k}\rangle d\mathbf{k} \right] \quad (2.31)$$

This way of obtaining the wavefunction has the advantage of being very descriptive, however it is not the way followed in this chapter. To get the dipole moment we use the time-evolution operators, that provide a rigorous framework

for understanding the quantum mechanics. In addition, we know which have been the approximations used in the dipole moment when Lewenstein model is used.

Using the Dirac notation instead of the wavefunction notation $\Psi(\mathbf{r}, t)$, where the wavefunction is related with the state vector $|\Psi(t)\rangle$ of the system at the time t by

$$\Psi(\mathbf{r}, t) \equiv \langle \mathbf{r} | \Psi(t) \rangle \quad (2.32)$$

The definition of the time-evolution operator in the Schrödinger picture $U(t, t_0)$ is such that

$$|\Psi(t)\rangle = U(t, t_0)|\Psi(t_0)\rangle \quad (2.33)$$

it indicates that when the time-evolution operator acts over the state $|\Psi(t_0)\rangle$ it generates a new state $|\Psi(t)\rangle$. The time-evolution operator satisfies the initial condition $U(t_0, t_0) = I$ where I is the unit operator, as a result

$$U(t, t_0)U(t_0, t) = I \rightarrow U^{-1}(t, t_0) = U(t_0, t) \quad (2.34)$$

On the other hand, when the operator evolution is applied to two consecutive states

$$U(t, t_0) = U(t, t_1)U(t_1, t_0) \quad (2.35)$$

The time-evolution operator requires satisfying the following expression to preserve the wavefunction norm

$$U^\dagger(t, t_0)U(t, t_0) = U(t, t_0)U^\dagger(t, t_0) = I \quad (2.36)$$

hence, the time-evolution operator is an unitary operator, then from above equations

$$U^\dagger(t, t_0) = U^{-1}(t_0, t) = U(t, t_0) \quad (2.37)$$

To see how the time-evolution operator depends on the Hamiltonian $H(t)$, we take the state vector $|\Psi(t)\rangle$ that is a solution of the TDSE.

$$i\frac{\partial}{\partial t}|\Psi(t)\rangle = H(t)|\Psi(t)\rangle \quad (2.38)$$

from Eq. (2.33), and replacing in above expression, the time-evolution operator satisfies

$$i\frac{\partial}{\partial t}U(t, t_0) = H(t)U(t, t_0) \quad (2.39)$$

where all terms are operators. Making use of the initial condition $U(t_0, t_0) = I$, the solution is given by the integral equation

$$U(t, t_0) = I - i \int_{t_0}^t H(t_1) U(t_1, t_0) dt_1 \quad (2.40)$$

Applying Eq. (2.37) to previous equation, a new second expression is obtained

$$U(t, t_0) = I - i \int_{t_0}^t U(t, t_1) H(t_1) dt_1 \quad (2.41)$$

Analytic expression of the time-evolution operator.

In some cases, it is possible to obtain an analytic expression for the time-evolution operator.

For a system field-free the time independent Hamiltonian H_0 has the eigenfunction $|\psi_k\rangle$ and E_k eigenvalues, that satisfies

$$H_0 |\psi_k\rangle = E_k |\psi_k\rangle \quad (2.42)$$

where any stationary solution in function of the eigenfunction could be written as

$$|\phi_k(t)\rangle = |\psi_k\rangle \exp(-iE_k t) \quad (2.43)$$

The Schrödinger equation is given by

$$i \frac{\partial}{\partial t} |\Psi_0(t)\rangle = H_0 |\Psi_0(t)\rangle \quad (2.44)$$

hence, a solution of above equation written as function of the eigenfunction has the form

$$|\Psi_0(t)\rangle = \sum_l a_k^0 |\phi_k(t)\rangle = \sum_k a_k^0 |\psi_k\rangle \exp(-iE_k t) \quad (2.45)$$

and writing the same state as function of the time-evolution operator

$$|\Psi_0(t)\rangle = U_0(t, t_0) |\Psi_0(t_0)\rangle \quad (2.46)$$

and replacing in the above Schrödinger in Eq. (2.44), the time-evolution operator corresponding to the Hamiltonian H_0 , satisfies

$$i \frac{\partial}{\partial t} U_0(t, t_0) = H_0 U_0(t, t_0) \quad (2.47)$$

where the solution in this case is simple compared with Eq. (2.40) because the H_0 is time independent

$$\begin{aligned} U_0(t, t_0) &= \exp[-iH_0(t - t_0)] \\ &= \sum_{k=0}^{\infty} \frac{1}{k!} (-i)^k H_0^k (t - t_0)^k \\ &= \sum_k \exp[-iE_k(t - t_0)] |\psi_k\rangle \langle \psi_k| \end{aligned} \quad (2.48)$$

where E_k is the eigenvalue, $|\psi_k\rangle$ is the eigenstate, the index k runs over all discrete and continuum states, and the matrix $|\psi_k\rangle \langle \psi_k|$ satisfies the closure relation

$$\sum_k |\psi_k\rangle \langle \psi_k| = I \quad (2.49)$$

The time-evolution operator from a free electron in the absence of a field can be written in term of plane-wave state $|\mathbf{k}\rangle$ as

$$U_{free}(t, t_0) = \int \exp[-iE_k(t - t_0)] |\mathbf{k}\rangle \langle \mathbf{k}| d\mathbf{k} \quad (2.50)$$

where E_k is the eigenstate of the equation $K|\mathbf{k}\rangle = E_k|\mathbf{k}\rangle$ whose value is $E_k = (k^2/2)$ the kinetic energy of a free particle.

The Gordon-Volkov time-evolution operator in the velocity gauge for free electrons under the interaction of the field using in Eq. (2.24) is given by

$$U_F^V = \int f_{\mathbf{k}}(t, t_0) |\mathbf{k}\rangle \langle \mathbf{k}| d\mathbf{k} \quad (2.51)$$

where $\mathbf{k}(\mathbf{r}) \equiv \langle \mathbf{r} | \mathbf{k} \rangle = (2\pi)^{-3/2} \exp(i\mathbf{k} \cdot \mathbf{r})$ and from the definition of the Gordon-Volkov state in Eq. (2.25) the U_F^V in atomic units is

$$U_F^V(t, t_0) = \int |\chi_{\mathbf{k}}^V(t)\rangle \langle \chi_{\mathbf{k}}^V(t_0)| \quad (2.52)$$

and in the gauge length

$$U_F^L(t, t_0) = \int |\chi_{\mathbf{k}}^L(t)\rangle \langle \chi_{\mathbf{k}}^L(t_0)| \quad (2.53)$$

A more general expression permit to write Gordon-Volkov state $|\chi_{\mathbf{k}_f}(t)\rangle$ as the state of the free particle $|\mathbf{k}\rangle$

$$|\chi_{\mathbf{k}_f}(t)\rangle = \exp \left[-i \int_{t_0}^t H_F(t') dt' \right] |\mathbf{k}_f\rangle \quad (2.54)$$

where $H_F(t) = K + H_{int}(t)$ denotes the Hamiltonian of a free particle under the interaction of a field.

The Gordon-Volkov wavefunction has decisive role in several applications. One of them is in the SFA theory, where is used to obtain the ionization rates calculated from the amplitude for the transition $\langle \chi(t) | \Psi(t) \rangle$.

So, any transition between an initial and final state has the following equation

$$a_{fi}(t) = \langle \phi_f(t) | \Psi_i(t) \rangle = \langle \phi_f(t) | U(t, t_0) \Psi_i(t_0) \rangle \quad (2.55)$$

and the probability of transition is given by

$$P_{if}(t) = a_{fi}(t) a_{fi}^*(t) = |a_{fi}(t)|^2 \quad (2.56)$$

and the transition rate is given by

$$W_{fi} = \frac{d}{dt} P_{fi}(t) \quad (2.57)$$

2.4.2 The perturbation theory.

In this section we develop the formalism of the time-evolution operator applied to the perturbation theory. Some of this results are used in the SFA theory in the following section,

In the perturbation theory, the electric field strength is much smaller than the atomic potential when both interact with the electrons. We can write the Hamiltonian of an atom under the interaction of an external field as

$$H(t) = H_0 + H_{int}(t) \quad (2.58)$$

here, H_0 is the field-free Hamiltonian that is time independent, and known as unperturbed part. $H_{int}(t)$ corresponds to the interaction part of the Hamiltonian that is time dependent. In the length gauge, the perturbed part is given by $H_{int}^L(t) = \mathbf{r} \cdot \mathbf{E}(t)$.

In this theory, in Eq. (2.58) is constituted by two parts completely independent. The first part H_0 , the eigenvalue and eigenstate is not affected by the presence of the electric field. Without this approximation, the energy level of the atom is modified when the atomic potential is superposed to external field, and as a consequence the wavefunction changes.

The interaction picture.

The interaction picture is an intermediate representation between the Schrödinger picture and the Heisenberg picture. In the Schrödinger picture, the state vector carry on the temporal dependence; in the Heisenberg picture, the temporal dependency is in the operators. The interaction picture is a suitable representation when the Hamiltonian is splitted in two parts: the time independent H_0 , and the time dependent $H_{int}(t)$.

The problem can be simplified by extracting the temporal dependency on $|\Psi(t)\rangle$ that corresponds to the H_0 , when $H_{int}(t)$ is an small perturbation. We now define

$$|\Psi_I(t)\rangle \equiv U_0^\dagger(t, t_0)|\Psi(t)\rangle = \exp[iH_0(t - t_0)]|\Psi(t)\rangle \quad (2.59)$$

where the state $|\Psi(t)\rangle$ corresponds to the Schrödinger picture, and $|\Psi_I(t)\rangle$ denotes the state in the interaction picture. At initial time t_0 when the field starts acting, $|\Psi_I(t_0)\rangle = |\Psi(t_0)\rangle$ and $H_{int}(t_0) = 0$. How the operator $U_0^\dagger(t, t_0)$ is unitary, the state $|\Psi_I(t)\rangle$ preserves some magnitudes as expectation value and the probability of the transition.

The transformation of the Schrödinger equation in the interaction picture gives the equation named as Tomonaga–Schwinger

$$i\frac{\partial}{\partial t}|\Psi_I(t)\rangle = H_{int}^I(t)|\Psi_I(t)\rangle \quad (2.60)$$

where from Eq. (2.37), (2.39), (2.58)

$$\begin{aligned} H_{int}^I(t) &= U_0^\dagger(t, t_0)H_{int}(t)U_0(t, t_0) \\ &= \exp[iH_0(t - t_0)]H_{int}(t)\exp[-iH_0(t - t_0)] \end{aligned} \quad (2.61)$$

A time-evolution operator in the interaction picture $U_I(t, t_0)$ is described by

$$|\Psi_I(t)\rangle = U_I(t, t_0)|\Psi_I(t_0)\rangle \quad (2.62)$$

and it satisfies the TDSE

$$i\frac{\partial}{\partial t}U_I(t, t_0) = H_{int}^I(t)U_I(t, t_0) \quad (2.63)$$

then, the equivalent expressions to (2.40) (2.41) are

$$U_I(t, t_0) = I - i \int_{t_0}^t H_{int}^I(t_1)U_I(t_1, t_0)dt_1 \quad (2.64)$$

and

$$U_I(t, t_0) = I - i \int_{t_0}^t U_I(t, t_1)H_{int}^I(t_1)dt_1 \quad (2.65)$$

The Schrödinger picture.

We will establish the relation between the Schrödinger picture and interaction picture with the end to have an expression of the time-evolution operator in the Schrödinger picture. From Eq. (2.62)

$$\begin{aligned}
|\Psi_I(t)\rangle &= U_0^\dagger(t, t_0)|\Psi(t)\rangle \\
&= U_0^\dagger(t, t_0)U(t, t_0)|\Psi(t_0)\rangle \\
&= U_0^\dagger(t, t_0)U(t, t_0)|\Psi_I(t_0)\rangle \\
&= U_I(t, t_0)|\Psi_I(t_0)\rangle
\end{aligned} \tag{2.66}$$

and from the properties of the time-evolution operator in Eq. (2.37)

$$\begin{aligned}
U_I(t, t_0) &= U_0^\dagger(t, t_0)U(t, t_0) \\
U(t, t_0) &= U_0(t, t_0)U_I(t, t_0)
\end{aligned} \tag{2.67}$$

since last equation indicates that the time-evolution operator in the Schrödinger picture is divided in two time-evolution operators: U_0 whose Hamiltonian is independent of the time, and U_I whose Hamiltonian is time dependent $H_{int}^I(t)$. Then, from Eqs. (2.64), (2.65), we get

$$\begin{aligned}
U(t, t_0) &= U_0(t, t_0) - i \int_{t_0}^t U_0(t, t_0)H_{int}^I(t_1)U_I(t_1, t_0)dt_1 \\
&= U_0(t, t_0) - i \int_{t_0}^t U_0(t, t_0)U_0^\dagger(t, t_1)H_{int}(t_1)U_0(t, t_1)U_0^\dagger(t_1, t_0)U(t_1, t_0)dt_1 \\
&= U_0(t, t_0) - i \int_{t_0}^t U_0(t, t_1)H_{int}(t_1)U(t_1, t_0)dt_1
\end{aligned} \tag{2.68}$$

and finally

$$U(t, t_0) = U_0(t, t_0) - i \int_{t_0}^t U(t, t_1)H_{int}(t_1)U_0(t_1, t_0)dt_1 \tag{2.69}$$

Above expressions are too complex to work them, but we can use an approximation based on the iterative solutions over $U(t, t_0)$. In the expression $U(t, t_0) = U_0(t, t_0)U_I(t, t_0)$, the iteration is only applied to the evolution operator in the interaction picture. Then, as first approximation is

$$U_I^{(0)}(t, t_0) = I \tag{2.70}$$

and replacing iteratively in Eqs. (2.64) or (2.65)

$$U_I^{(n)}(t, t_0) = I - i \int_{t_0}^t H_{int}^I(t_1) U_I^{(n-1)}(t_1, t_0) dt_1 \quad (2.71)$$

therefore, the final solution is

$$U_I(t, t_0) = \sum_{n=0}^{\infty} \hat{U}_I^{(n)}(t, t_0) \quad (2.72)$$

since every term of above summation corresponds to

$$\hat{U}_I^{(n)}(t, t_0) = (-i)^n \int_{t_0}^t dt_1 \int_{t_0}^{t_1} dt_2 \dots \int_{t_0}^{t_{n-1}} dt_n H_{int}^I(t_1) H_{int}^I(t_2) \dots H_{int}^I(t_n) \quad (2.73)$$

this equation requires the following ordering in the time $t \geq t_1 \geq t_2 \geq \dots \geq t_n \geq t_0$, also Hamiltonian inside the integral does not commute when $t_i \neq t_j$

An approximated solution of the time-evolution operator in the Schrödinger picture is obtained from Eqs. (2.68) or (2.69). Then, replacing Eq. (2.61), (2.72) in Eqs. (2.68) or (2.69), the time-evolution operator is

$$U(t, t_0) = U_0(t, t_0) + \sum_{n=0}^{\infty} \hat{U}^{(n)}(t, t_0) \quad (2.74)$$

where $\hat{U}^{(n)}$ for $n \geq 1$ is given by

$$\begin{aligned} \hat{U}^{(n)}(t, t_0) = & (-i)^n \int_{t_0}^t dt_1 \int_{t_0}^{t_1} dt_2 \dots \int_{t_0}^{t_{n-1}} dt_n U_0(t, t_1) H_{int}(t_1) \\ & \times U_0(t_1, t_2) H_{int}(t_2) \dots U_0(t_{n-1}, t_n) H_{int}(t_n) U_0(t_n, t_0) \end{aligned} \quad (2.75)$$

Since the state evolution from the initial state $|\Psi(t_0)\rangle$ becomes

$$|\Psi(t)\rangle = U_0(t, t_0) |\Psi(t_0)\rangle + \sum_{n=0}^{\infty} \hat{U}^{(n)}(t, t_0) |\Psi(t_0)\rangle \quad (2.76)$$

Then, the final state is obtained as sequence of successive approximations. When $\hat{U}^{(n)}(t, t_0)$ acts on $|\Psi(t_0)\rangle$ can be interpreted as, the state $|\Psi(t_0)\rangle$ evolves freely without the field interaction from the initial time t_0 to t_n , at time t_n interacts with the laser and then the new state evolves again freely until the time t_{n+1} where interacts with the field, and so on. In the dipole approximation, in each interaction with the laser, the electron absorbs or emits a photon.

2.4.3 The strong-field approximation.

The Hamiltonian equation under the single-active electron approximation is given by

$$H(t) = H_0 + H_{int}(t) \quad (2.77)$$

where H_0 denotes the Hamiltonian field-free and $H_{int}(t)$ describes the interaction of the electric field with the single active electron. Writing the Hamiltonian of a free electron H_F under the interaction of an electric field as

$$H_F(t) = H(t) - V = K - H_{int}(t) \quad (2.78)$$

where V is the atomic potential operator, and K is the kinetic energy operator, defined as $K = \mathbf{p}^2/2$.

Any state $|\phi(t_0)\rangle$ at the initial t_0 time can be written as a function of the atomic states $|\psi_0\rangle$ from the atomic Hamiltonian H_0 as $|\phi(t_0)\rangle = |\psi_0\rangle \exp(-I_p t_0 i)$, where I_p is the eigenvalue of eigenstate $|\psi_0\rangle$ from the Hamiltonian operator H_0 . Therefore, a solution using the full Hamiltonian in Eq. (2.38) can be obtained using anyone of Eqs. (2.68), (2.69)

$$|\psi(t)\rangle = |\phi(t_0)\rangle - i \int_{t_0}^t U(t, t') H_{int}(t') |\phi_0(t')\rangle dt' \quad (2.79)$$

where the expression for $U(t, t')$ is difficult to obtain. To this end, we follow similar steps we did in the section 2.4.2 although changing the role of some operators. In the above equation the time-evolution operator acts on a state after interacting with the external lasers by means of the absorption or emission a photon. In this new frame, the electron moves freely in the electric field, and punctually interacts with the atom. Then, the role of the perturbation rest on atomic potential. In Eq. (2.68) changing $H_{int}^I \rightarrow V$, and $U_0 \rightarrow U_F$, we get

$$U(t, t') = U_F(t, t') - i \int_{t'}^t U_F(t, t_1) V U(t_1, t') dt_1 \quad (2.80)$$

and

$$U(t, t') = U_F(t, t') - i \int_{t'}^t U(t, t_1) V U_F(t_1, t') dt_1 \quad (2.81)$$

where $U_F(t, t_0)$ is known as Gordon-Volkov time-evolution operator and is based on the expression of the free-electron wavefunction in an electric field $|\chi_k(t)\rangle$

[51]

$$\begin{aligned}
 U_F(t, t') &= \int |\chi_{\mathbf{k}}(t)\rangle \langle \chi_{\mathbf{k}}(t')| dk \\
 |\chi_{\mathbf{k}}(t)\rangle &= |\mathbf{k}\rangle e^{-i \int_{t'}^t H_F(t'') dt''}
 \end{aligned} \tag{2.82}$$

A series expansion of the time-evolution operator is

$$U(t, t') = U_F(t, t') + \sum_{n=1}^{\infty} \hat{U}^{(n)}(t, t') \tag{2.83}$$

where $\hat{U}^{(0)} = U_F(t, t')$, $\hat{U}^{(n)}$ for $n \geq 1$ is given by

$$\begin{aligned}
 \hat{U}^{(n)}(t, t') &= (-i)^n \int_{t_0}^t dt_1 \int_{t'}^{t_1} dt_2 \dots \int_{t_0}^{t_{n-1}} dt_n U_F(t, t_1) V \\
 &\quad \times U_F(t_1, t_2) V(t_2) \dots U_F(t_{n-1}, t_n) V(t_n) U_F(t_n, t')
 \end{aligned} \tag{2.84}$$

So, replacing the time-evolution operator in Eqs. (2.83), (2.79)

$$|\Psi(t)\rangle = |\phi_0(t)\rangle + \sum_{n=0}^{\infty} |\hat{\Psi}^{(n)}(t)\rangle \tag{2.85}$$

where

$$|\hat{\Psi}^{(n)}(t)\rangle = -i \int_{t_0}^t dt' \hat{U}^{(n)}(t, t') H_{int}(t') |\phi_0(t')\rangle \tag{2.86}$$

and writing the state as addition of several corrections:

$$|\Psi^{(n)}(t)\rangle = |\phi_0(t)\rangle + \sum_{j=0}^n |\hat{\Psi}^{(j)}(t)\rangle \tag{2.87}$$

Eq. (2.87) can be interpreted as the addition of several states.

The approximation of the first order, $n = 0$, is the addition of the ground state and the free electron. The second term on the right-hand equation indicates that an electron in the atom absorbs a photon at time t' , as a result, the electron travels as free particle in the electric field until the final time t .

$$|\Psi^{(0)}(t)\rangle = |\phi_0(t)\rangle - i \int_{t_0}^t dt' U_F(t, t') H_{int}(t') |\phi_0(t')\rangle \tag{2.88}$$

In this case, the electron reaches the continuum directly, without any intermediate state.

The approximation of second order , $n = 1$, is given by

$$\begin{aligned} |\Psi^{(1)}(t)\rangle &= |\phi_0(t)\rangle - i \int_{t_0}^t dt' U_F(t, t') H_{int}(t') |\phi_0(t')\rangle \\ &\quad - \int_{t_0}^t dt' \int_{t_0}^{t'} dt'' U_F(t, t'') V U_F(t'', t') H_{int}(t') |\phi_0(t')\rangle \end{aligned} \quad (2.89)$$

the last integral describes an electron is ionized at time t' , it travels freely in the electric field until time t'' when the electron collides with the atom to continue as free electron in the field until the current time t . How the SFA is based on single-electron approximation all recollision with the parent ion are elastic because there is not excited internal states. For the atoms with more than one electron, the recollision can be elastic or inelastic.

2.4.4 The dipole moment.

One of the possible mathematical expressions of the dipole moment is given by [33]

$$\mathbf{d}(t) = -\langle \Psi(t) | \mathbf{r} | \Psi(t) \rangle \quad (2.90)$$

where the minus sign comes from the charge of the electron. The dipole module can be easily calculated when we know the analytical expression of $|\Psi(t)\rangle$. Unfortunately, this is extremely difficult; so, we use the approximation from the last section. Based on the first order approximation of the state $|\Psi(t)\rangle$

$$\begin{aligned} \mathbf{d}(t) &= -\langle \Psi^{(0)}(t) | \mathbf{r} | \Psi^{(0)}(t) \rangle \\ &= -[\langle \phi_0(t) | \mathbf{r} | \phi_0(t) \rangle + 2\Im m \int_{t_0}^t dt' \langle \phi_0(t) | \mathbf{r} U_F(t, t') H_{int}(t') | \phi_0(t') \rangle \\ &\quad + \int_{t_0}^t dt' \int_{t_0}^{t'} dt'' \langle \phi_0(t') | H_{int}^\dagger(t') U_F(t', t) \mathbf{r} U_F(t, t'') H_{int}(t'') | \phi_0(t'') \rangle] \end{aligned} \quad (2.91)$$

The first term of the right-hand side is vanished because $\mathbf{r}$ is an operator with odd parity. The third term is the continuum-continuum transitions and its contribution is insignificant compared to the second term. So, the dipole moment is finally given by

$$\mathbf{d}(t) = i \int_{t_0}^t dt' \langle \phi_0(t) | \mathbf{r} | U_F(t, t') H_{int}(t') \phi_0(t') \rangle + cc \quad (2.92)$$

The high-order approximations permit to study other processes. In this way, the second order approximation $\Psi^{(1)}(t)$ includes the collisional process between the electron and the parent ion.

The evolution of the wavefunction in the ground state $|\psi_0\rangle$ with the ionization potential I_p , is given by

$$|\phi_0(t)\rangle = |\psi_0\rangle e^{-iI_p t} \quad (2.93)$$

and the Gordon-Volkov propagator U_F (2.82)

$$\begin{aligned} U_F(t, t_0) &= \int |\chi_{\mathbf{k}}(t)\rangle \langle \chi_{\mathbf{k}}(t_0)| dk \\ |\chi_{\mathbf{k}}(t)\rangle &= |\mathbf{k}\rangle e^{-i \int_{t_0}^t H_F(t') dt'} \end{aligned} \quad (2.94)$$

In the length gauge, the Lewenstein expression of the dipole moment is [64]

$$\begin{aligned} \mathbf{d}(t_r) &= i \int_{t_0}^{t_r} dt_i \int d\mathbf{k} \langle \Psi_0 | \mathbf{r} | \mathbf{v}(\mathbf{k}, t_r) \rangle \\ &\quad \langle \mathbf{v}(\mathbf{k}, t_i) | \mathbf{E}(t_i) \cdot \mathbf{r} | \Psi_0 \rangle e^{-iS(\mathbf{k}, t_i, t_r)} + cc \end{aligned} \quad (2.95)$$

where t_r is the recombination time, t_i is the ionization time, $H_{int}(t) = \mathbf{E}(t) \cdot \mathbf{r}$ is the interaction Hamiltonian in the length gauge, and $\mathbf{v}(\mathbf{k}, t)$ is the kinetic momentum that is related with the canonical momentum $\mathbf{k}$ in the length gauge by the expression $\mathbf{v}(\mathbf{k}, t) = \mathbf{k} + \mathbf{A}(t)$ where $\mathbf{A}(t)$ denotes the vector potential. $S(\mathbf{k}, t_i, t_r)$ is the quasi-classical action that describes the motion of an electron in an electric field.

$$S(\mathbf{k}, t_i, t_r) = \int_{t_i}^{t_r} \frac{[\mathbf{v}(\mathbf{k}, t)]^2}{2} + I_p dt \quad (2.96)$$

The master piece of the Lewenstein model is the dipolar transition matrix $\langle \mathbf{k} | \mathbf{r} | \Psi_0 \rangle$. The following steps are oriented to obtain its analytical expression. To this end, we use the mathematical formula of wavefunction in the momentum space $\langle \mathbf{k} | \Psi_0 \rangle$ given by

$$\begin{aligned} \langle \mathbf{k} | \Psi_0 \rangle &= (2\pi)^{-3/2} \int \langle \mathbf{r} | \Psi_0 \rangle e^{-i\mathbf{k} \cdot \mathbf{r}} d\mathbf{r} \\ &= (2\pi)^{-3/2} \int \Psi_0(\mathbf{r}) e^{-i\mathbf{k} \cdot \mathbf{r}} d\mathbf{r} \end{aligned} \quad (2.97)$$

where this equation is the Fourier transform of the $\Psi_0(\mathbf{r}) \equiv \langle \mathbf{r} | \Psi_0 \rangle$. Then, $\langle \mathbf{k} | \mathbf{r} | \Psi_0 \rangle$ is the derivative of the $\langle \mathbf{k} | \Psi_0 \rangle$. The complex exponential in the integral corresponds to the wavefunction of a free electron $|\mathbf{k}\rangle$ [27]

$$\langle \mathbf{k} | \mathbf{r} | \Psi_0 \rangle = i \nabla_{\mathbf{k}} \langle \mathbf{k} | \Psi_0 \rangle \quad (2.98)$$

here $\nabla_{\mathbf{k}}$ is the gradient function in the momentum space $\mathbf{k} = (k_x, k_y, k_z)$.

The dipolar transition matrix $\langle \mathbf{k} | \mathbf{r} | \Psi_0 \rangle$ has different equations for different the ground state equations $|\Psi_0\rangle$: Gaussian functions [64], Slater-type orbitals (STO) [44], asymptotic wavefunctions [51], [41].

2.4.5 The wavefunction in the momentum space.

The first step is to obtain the wavefunction in the momentum space, and later to calculate its derivative respect to $\mathbf{k}$. The wavefunction of the electron in the ground state can be splitted in two the radial and the angular part.

$$\Psi_{nlm}(r, \theta, \phi) = R_{nl}(r) Y_l^m(\theta, \phi), \quad R_{nl}(r) = \frac{P_{n,l}(r)}{r} \quad (2.99)$$

The radial component of the wavefunction based on the Slater orbital is

$$P_{n,l}(r) = 2^{l+1} \sqrt{\frac{(n-l-1)!}{n(n+l)!}} \kappa^{l+3/2} \exp(-\kappa r) r^{l+1} L_{n-l-1}^{2l+1}(2r\kappa) \quad (2.100)$$

where L_{n-l-1}^{2l+1} are the Laguerre polynomials, and $\kappa = \sqrt{2I_p}$ is related with the ionization potential.

By means of the Fourier transform (2.97), we obtain the wavefunction in the momentum space.

$$\begin{aligned} \mathcal{F}[R_{nl}(r) Y_l^m(\theta, \phi)] &= (2\pi)^{-3/2} \int e^{-i\mathbf{k}\cdot\mathbf{r}} R_{nl}(r) Y_l^m(\theta, \phi) d\mathbf{r} \\ &= F_{nl}(k) Y_l^m(\theta, \phi) \end{aligned} \quad (2.101)$$

where the complex exponential can be decomposed in spherical harmonics as

$$e^{i\mathbf{k}\cdot\mathbf{r}} = 4\pi \sum_{l=0}^{\infty} i^l j_l(kr) \sum_{m=-l}^{m=l} Y_l^{m*}(k, \theta', \phi') Y_l^m(r, \theta, \phi) \quad (2.102)$$

and using the coupling rules from the spherical harmonics, the radial part in the momentum space is given by

$$F_{nl}(k) = -i^l \sqrt{\frac{2}{\pi}} \int_0^{\infty} r^2 j_l(kr) R_{nl}(r) dr \quad (2.103)$$

here, k is the modulus of the momentum, and $j_l(kr)$ is the Bessel function. Then, the wavefunction of the Slater-type orbitals in the momentum space:

$$\langle \mathbf{k} | \Psi_0 \rangle = F_{nl}(k) Y_l^m(\theta, \phi) \quad [44]$$

$$F_{nl}(k) = \sqrt{\frac{2n(n-l-1)!}{\pi(n+l)!}} 2^{2l+2} l! \kappa^{-\frac{3}{2}} \left(\frac{k}{\kappa}\right)^l \times \frac{1}{((k/\kappa)^2 + 1)^{l+2}} C_{n-l-1}^{(l+1)} \left(\frac{(k/\kappa)^2 - 1}{(k/\kappa)^2 + 1}\right) \quad (2.104)$$

where $C_{n-l-1}^{(l+1)}$ are the Gegenbauer polynomials. [4]

For a hydrogenic atom with the quantum numbers $n = 1, l = 0, m = 0$

$$\langle \mathbf{k} | \Psi_0 \rangle = \frac{2^{15/4} I_p^{5/4}}{\sqrt{\pi}(k^2 + 2I_p)^2} \frac{1}{\sqrt{4\pi}} \quad (2.105)$$

and from Eq. (2.98), the dipolar transition matrix is given by

$$\langle \mathbf{k} | \mathbf{r} | \Psi_0 \rangle = -i \frac{2^{23/4} I_p^{5/4} \mathbf{k}}{\sqrt{\pi}(k^2 + 2I_p)^3} \frac{1}{\sqrt{4\pi}} \quad (2.106)$$

This expression is very extended in the literature, although rarely times is indicated how it is obtained.

The second mathematical model is based on the asymptotic part of the wavefunction whose the radial part is the following

$$R_{n^*,l} \rightarrow C_{asym}(r) = C_{asym} \kappa^{3/2} (\kappa r)^{Z_c/\kappa-1} \exp(-\kappa r), r \rightarrow \infty \quad (2.107)$$

where n^* is the effective principal quantum number with $I_p = Z_c^2/(2n^{*2})$, C_{asym} is the asymptotic coefficient, Z_c is effective nuclear charge, and C_{asym} is given by

$$C_{asym} = \left[\frac{2^{2n}}{n(n+l)!(n-l-1)!} \right]^{1/2} \quad (2.108)$$

For hydrogenic atoms in the ground state $C_{asym} = 2, Z_c = \kappa = Z, l = m = 0$ and after applying the Fourier transform [41].

$$\langle \mathbf{k} | \Psi_0 \rangle \simeq \xi_l C_{asym} \frac{\Gamma(1 + Z_c/\kappa)}{\sqrt{2\pi^2 \kappa^3}} \left(\frac{2\kappa^2}{k^2 + \kappa^2} \right)^{Z_c/\kappa+1} Y_l^m(\theta, \phi) \quad (2.109)$$

where Γ denotes the Euler's gamma function, l and m are the quantum numbers, and $\xi_l = 1$ for $k \rightarrow i\kappa$, $\xi_l = -1^l$ for $k \rightarrow -i\kappa$, it depends on the way is

approximated to the pole of the function in the complex plane. For the hydrogenic atoms with $n = 1, l = 0$ has the same expression as Eq. (2.105), although it is different for other atoms like the neon.

Finally, the third model is based on the Gaussian wavefunction, which equation in the momentum space for hydrogenic atoms is given by [64]

$$\langle \mathbf{k} | \mathbf{r} | \Psi_0 \rangle = i \left(\frac{1}{\pi\alpha} \right)^{3/4} \frac{\mathbf{k}}{\alpha} \exp \left(\frac{-\mathbf{k}^2}{2\alpha} \right), \alpha = 0.8I_p \quad (2.110)$$

2.4.6 The saddle point approximation.

A solution of the integral equation (2.95) is possible to get by means of the stationary phase approximation. The method applies to the oscillatory integrals with complex exponent. The main idea of the stationary phase methods relies on the cancellation of sinusoids with rapidly varying phase. The solution of the integral is a point where the phase varies slowly, that is, where the derivative of the complex exponential is zero. Therefore, if we get the derivative of the action over all its variables and equate it to zero, we have a set of equations that must be solved simultaneously:

$$\nabla_{\mathbf{k}} S(\mathbf{k}, t_i, t_r) = (t_r - t_i)\mathbf{k} + \int_{t_i}^{t_r} \mathbf{A}(t) dt = 0 \quad (2.111)$$

$$\frac{\partial S(\mathbf{k}, t_i, t_r)}{\partial t_i} = \frac{[\mathbf{v}(\mathbf{k}, t_i)]^2}{2} + I_p = 0 \quad (2.112)$$

$$\frac{\partial S(\mathbf{k}, t_i, t_r)}{\partial t_r} = \frac{[\mathbf{v}(\mathbf{k}, t_r)]^2}{2} + I_p - \omega = 0 \quad (2.113)$$

The numerical solution is not simple, it includes an integral of the potential vector, that only has solution for a simple cases. Kovacs and Tosa [58] they establish a procedure to extend the use of the above equation for the cases in which the potential vector would be of any form by means of Taylor expansions.

The numerical solutions of equations (2.111) - (2.113) produce a set of the complex solutions $(\mathbf{k}_s, \tau_r, \tau_i)$ named saddle points. This points identify a set of the electron trajectories that due to its contribution to the generation of the harmonics are the most important trajectories. Thus, once the trajectories are identified, the dipole moment can be described by a small set of trajectories, two

per subcycle. Here, $\mathbf{k}_s$ is the complex momentum, τ_i is the complex ionization time, and τ_r is the complex recombination time.

Eq. (2.111) indicates the momentum conservation $\mathbf{k}_s$. Eq. (2.112) calculates the ionization time when the kinetic and potential energy has the same value. Eq. (2.113) calculates the recombination time when the emitted photon ω is the addition of the kinetic and potential energy.

Figure 2.1: Fig. (a) shows the modulus of dipole moment per harmonic. The dotted line corresponds to short trajectories, the continuous lines is for long trajectories, and the thin lines represent the not convergent trajectories. Fig. (b) shows the dependency on the emitted photon frequency with the ionization and recombination times. The blue lines indicate the frequency of the photon emitted at the recombination time. The green lines indicates that the frequency of the emitted photon depends on the ionization time. The dotted line represents the short trajectories, and the continuous line is for the long trajectories. Both blue lines converge at the cutoff frequency. The number indicates in which visit to the parent ion the electron is recombined.

Based on the excursion time, the solutions are classified as long and short see Fig. 1.5. For the long paths, the electrons are set free near the maximum

value of the electric field and its excursion time in the continuum is around $\Re[\tau_r - \tau_i] \sim T$, where T is the electric field period. On the contrary, the short paths correspond to the electrons are set free at lower intensities of electric field with an excursion time given by $\Re[\tau_r - \tau_i] \sim T/2$ and the electrons return to parent ion earlier than in the long paths. Some times the electron does not recombines in the first visit to the parent ion, when the sign of the electric field changes, the electron moves to its origin. The electron can recombine in the second or third visit to the ion.

Fig. 2.1 shows the spectrum of the neon from a couple of short and long trajectories using a laser with wavelength $\lambda = 800 \text{ nm}$, and with the intensity $3.0 \times 10^{14} \text{ W/cm}^2$. This spectrum is produced by the electrons that are ionized in a single subcycle. In this case for those electrons that are set free near the time of 440 au . see green lines at Fig. 2.1(b). The summation of the contributions from all subcycles produces a typical spectrum as of Fig. 2.3, that is the result of the interference between all subcycles.

Fig. 2.1(b) exhibits three blue-line bells. Each blue bell describes how it relates to the frequency emitted by a photon (abscissa axis) with the recombination time (ordinate axis). The number indicates in which visit to the parent ion the recombination occurs. There are three other green bells, by the times of ionization, although it is not possible to distinguish in Fig. .

For the curves corresponding to the first visit to the parent ion, the modulus of the short trajectories has much lower value than of the long trajectories up to the cutoff where both trajectories converges. For the other two curves, the differences between the modules for short and long trajectories are not so great, although in both cases short trajectories have a higher dipolar modulus. The module differences between paths are used in the following chapters to simplify some calculations, taking only those paths with the greater module.

Every bell from Fig. 2.1(b) has its equivalent curve in Fig. 2.1(a). In this way, from both figures we have ionization and recombination times together intensity and frequency of the harmonics.

Fig. 2.2 shows the phase of the harmonics for the short and long trajectories. The phase is a crucial magnitude in the phase-matching calculations, to improve the intensity of the harmonics. The first direct measurements of the atomic dipole phase in the process of high-order harmonic generation was done by Corsi et al.[32], although they can not separate the phase of the long from the short trajectories for the harmonics in the plateau.

The contribution to the intensity of the harmonics is reduced when the elec-

Figure 2.2: Figure shows how the phase of the dipole moment phase changes with the harmonic of the photon emitted at the recombination time. The dotted lines represent the short trajectories and the continues lines represent the long trajectories.

tron needs several visits to the parent ion until finally it recombines. This is because the attenuation factor $2\pi/(i(\tau_r - \tau_i))$ from (2.115) decreases when increases the recombination time τ_r . As a result, we limit to three visits $\Re[\tau_r - \tau_i] < 3T$ all calculations.

The Fourier transform of the first term of Eq. (2.92)

$$\mathbf{d}(\omega) = i \int_{-\infty}^{\infty} dt_r \int_{t_0}^{t_r} dt_i \int d\mathbf{k} \langle \Psi_0 | \mathbf{r} | \mathbf{v}(\mathbf{k}, t_r) \rangle \langle \mathbf{v}(\mathbf{k}, t_i) | \mathbf{E}(t_i) \cdot \mathbf{r} | \Psi_0 \rangle e^{i(\omega t_r - S(\mathbf{k}, t_i, t_r))} \quad (2.114)$$

where the complex conjugate part is not included because it does not produce a valid results for the positive frequencies. The Fourier transform of a function produces negative and positive frequencies.

The integration of equation (2.114) by means of stationary phase approximation, gives as a result a superposition of the quantum paths

$$\begin{aligned} \mathbf{d}(\omega) = \sum_q \left(\frac{2\pi}{i(\tau_{qr} - \tau_{qi})} \right)^{3/2} C_x \\ \times \langle \mathbf{v}(\mathbf{k}_{qs}, \tau_{qr}) | \mathbf{r} | \Psi_0 \rangle^* \times \langle \mathbf{v}(\mathbf{k}_{qs}, \tau_{qi}) | \mathbf{E}(t_i) \cdot \mathbf{r} | \Psi_0 \rangle \\ \times e^{i(\omega \tau_{qr} - S(\mathbf{k}_{qs}, \tau_{qi}, \tau_{qr}))} \end{aligned} \quad (2.115)$$

where the index of the quantum path q identifies a set of values $(\mathbf{k}_{qs}, \tau_{qr}, \tau_{qi})$ which are the solutions of Eqs.(2.111) -(2.113). C_x has different values depending on the expression taken for the dipolar transition matrix.

When, the dipolar transition matrix comes from a STO or asymptotic wavefunctions has a singularity in the ionization term. This is because the solution

of Eq. (2.112) produces a singularity in the equation (2.106). To overcome this, in the integration of Eq. (2.114) we use the approximation [41]:

$$\int dt \frac{\exp[-\lambda f(t)]}{(t - t_s)^\nu} \simeq i^\nu \sqrt{\pi} \frac{\Gamma(\nu/2)}{\Gamma(\nu)} [2\lambda f''(t_s)]^{(\nu-1)/2} e^{-\lambda f(t_s)} \quad (2.116)$$

where λ is large, $f(t)$ is a slowly varying function that could be approximated by

$$f(t) \simeq f(t_s) + f'(t_s)(t - t_s) + 1/2 f''(t_s)(t - t_s)^2 + .. \quad (2.117)$$

with $f'(t_s) = 0$ and $f''(t_s) \neq 0$, and $\Gamma(\nu)$ denotes the gamma function.

The dipolar transition matrix from the Gaussian wavefunction in Eq. (2.110) is free of the singularity problem. Then, the coefficient C_x for the Gaussian wavefunction is given by

$$C_x = \frac{2\pi}{\sqrt{\det S''}} \quad (2.118)$$

where the $\det S''$ comes from the simultaneous integration with both variables τ_i, τ_r [76].

For the STO and asymptotic wavefunctions, the C_x coefficient is

$$C_x = \sqrt{-2\pi i} \left(\frac{\partial^2 S}{\partial t_r^2} \right)_{\tau_{qr}}^{-1/2} \quad (2.119)$$

Due to the presence of the singularity at τ_i implies the integration had been solved sequentially in the variables τ_i and τ_r . This is the reason because there are different expressions for C_x .

Finally, the determinant given by

$$\det S'' = \left(\frac{\partial^2 S}{\partial t_r \partial t_i} \right)^2 - \left(\frac{\partial^2 S}{\partial t_r^2} \right) \left(\frac{\partial^2 S}{\partial t_i^2} \right) \quad (2.120)$$

The numerical values of the second derivative of action are given by:

$$\begin{aligned} \left(\frac{\partial^2 S}{\partial t_r^2} \right)_{\tau_{qr}} &= -\frac{2(\omega - I_p)}{\tau_{qr} - \tau_{qi}} - \mathbf{E}(\tau_{qr}) \cdot \mathbf{v}(\mathbf{k}_{qs}, \tau_{qr}) \\ \left(\frac{\partial^2 S}{\partial t_i^2} \right)_{\tau_{qi}} &= \frac{2I_p}{\tau_{qr} - \tau_{qi}} + \mathbf{E}(\tau_{qi}) \cdot \mathbf{v}(\mathbf{k}_{qs}, \tau_{qi}) \\ \left(\frac{\partial^2 S}{\partial t_r \partial t_i} \right)_{\tau_{qr}\tau_{qi}} &= \frac{\mathbf{v}(\mathbf{k}_{qs}, \tau_{qi}) \cdot \mathbf{v}(\mathbf{k}_{qs}, \tau_{qr})}{\tau_{qr} - \tau_{qi}} \end{aligned} \quad (2.121)$$

Integration by parts: case dipolar matrix based on STO.

The dependency on the electric field could be removed with the method of integration by parts [73] [51], by the expression

$$\langle \mathbf{k} | H_{int}(t) | \Psi_0 \rangle = -id/dt \langle \mathbf{k} | \Psi_0 \rangle \quad (2.122)$$

using the conditions that $H_{int}(t)$ is zero at the initial and end times of the laser pulse. This procedure only is explained for the dipolar transition matrix based on Slater-type orbital. For Gaussian wavefunction have a similar development. The integration by parts in Eq. (2.115) gives

$$\begin{aligned} \mathbf{d}(\omega) = & \sum_q \left(\frac{2\pi}{i(\tau_{qr} - \tau_{qi})} \right)^{3/2} \sqrt{-2\pi i} \times \left(\frac{\partial^2 S}{\partial t_r^2} \right)^{-1/2}_{\tau_{qr}} \\ & \times e^{i(\omega\tau_{qr} - S(\mathbf{k}_{qs}, \tau_{qi}, \tau_{qr}))} \\ & \times \langle \Psi_0 | \mathbf{r} | \mathbf{v}(\mathbf{k}_{qs}, \tau_{qr}) \rangle d_{ion}(q) \end{aligned} \quad (2.123)$$

where the electric field has been removed. The term $d_{ion}(q)$ has expression

$$d_{ion}(q) = (-2)^{-\nu} M(\mathbf{v}(\mathbf{k}_{qs}, \tau_{qi})) J(\nu - 1, S) \left(\frac{\partial^2 S}{\partial t_i^2} \right)^{1-\nu}_{\tau_{qi}} \quad (2.124)$$

where $J(\nu - 1, S)$ contains the approximation from Eqs. (2.116), (2.117)

$$J(\nu, S) = i^\nu \sqrt{\pi} \frac{\Gamma(\nu/2)}{\Gamma(\nu)} \left(2i \left(\frac{\partial^2 S}{\partial t_i^2} \right)_{\tau_{qi}} \right)^{(\nu-1)/2} \quad (2.125)$$

Finally, $M(\mathbf{v})$ and ν come of splitting $\langle \mathbf{v} | \Psi_0 \rangle$ in two parts: the coefficient $1/(v^2 + 2I_p)^\nu$ and the rest, then $M(\mathbf{v})$ and ν are

$$\langle \mathbf{v} | \Psi_0 \rangle = \frac{M(\mathbf{v})}{(v^2 + 2I_p)^\nu} \quad (2.126)$$

and for hydrogenic atom from Eq. (2.105)

$$\nu = 2, \quad M(\mathbf{v}) = \frac{2^{15/4} I_p^{5/4}}{\sqrt{\pi}} Y_0^0, \quad Y_0^0 = \frac{1}{\sqrt{4\pi}} \quad (2.127)$$

where Y_0^0 is the spherical harmonic $l = 0$ and $m = 0$ from the 1s level.

Figure 2.3: Figure shows the comparison between two spectra. The black line corresponds to a spectrum calculated with the Lewenstein model. Red line corresponds to a spectrum calculated using the saddle points approximation. The spectra was generated with a laser with a central wavelength of $\lambda = 800 \text{ nm}$, and with the intensity $3.0 \times 10^{14} \text{ W/cm}^2$ for a neon atom using the dipolar transition matrix given by Eq. (2.106)

The success of the saddle point approximation can be confirmed with the comparison in Fig. 2.3. Both curves present similar values from the harmonic 17^{th} onward. For lower-order harmonics there are no data, because the current implementation in the program only works with photons whose energies are greater than I_p . O. Smirnova and M Ivanov [76] indicate how to extend the spectrum in lower frequencies than I_p .

An example of the possibilities of this approximation is plotted in Fig. 2.4. This spectrum is obtained using a couple of trajectories from a single subcycle in the region where the envelope of the laser pulse is constant. The atom used is the neon. The laser pulse contains 18 cycles of which 2 cycles are ascending, another 2 cycles descending and 14 with constant intensity.

Fig. 2.4 depicts how the cutoff changes from the 65^{th} harmonic with an intensity of $4.0 \times 10^{14} \text{ W/cm}^2$ to the 53^{th} harmonic with an intensity $3.0 \times 10^{14} \text{ W/cm}^2$. Although the most impressive result is the variation of the dipole modulus with the laser intensity. Fig. 2.4 shows that the value of the harmonics intensity oscillates with the lasers intensity. These changes present a periodic pattern, although not the same for all harmonics. The oscillations for an specific harmonic is the result of the interference of the short and long trajectories of a single subcycle.

T Auguste et al.[5] perform a study on the dependence of the intensity of

the harmonics with the frequency of the laser. Using a Gaussian model, they found that $|d(\omega)|^2$ depends on ω^3 through out the dispersion factor from Eq. (2.115) and from the imaginary part of the action.

$$\begin{aligned} |d(\omega)|^2 &= \propto \omega^3 \exp(\Im m[S]) \propto \omega^3 \exp \left[-\frac{2(2I_p)^{3/2}}{3E_0} \right] \\ &= \omega^3 \exp \left[-\frac{4\gamma I_p}{3\omega} \right]; \quad \gamma = \frac{\omega \sqrt{2I_p}}{E(t_i)} \end{aligned} \quad (2.128)$$

where the exponential is related with the ionization rates (2.143). And the Schiessl et al. [89] carried out a similar study with the laser frequency although paying attention to combine intensity from a range of harmonics.

Figure 2.4: Figure shows how the modulus of the dipole moment changes when the laser intensity is in the range of $3.0 - 4.0 \times 10^{14} \text{ W/cm}^2$.

The uniform approximation.

The saddle point approximation fails for the harmonics near the cutoff when both short and long saddle points are close each other. There is a critical point where one of the two solutions must be discarded because it diverges, see the thin blue lines in Fig. 2.1(a). This problem was initially tackled eliminating those points where the trajectory diverges. As a result, in the spectrum appears jumps that are not in correspondence to the expected smooth behavior.

When both saddle points are close to each other, both solutions are not independent and should be treated as a whole. The solution used here is based on the Stokes transitions [22] [21] and is obtained through Airy functions [90].

The critical point ω_c where takes place the transition is named anti-Stokes transition and is obtained when the following condition is fulfilled

$$\Im m S_j = \Im m S_i \quad (2.129)$$

where S_i and S_j are the action of each trajectory, usually the short and long electron trajectories.

To get over this problem, both saddle points is solved simultaneously, instead separately as occurred in the previous section [23] [100]. The integral must be solved has the following form [36]

$$M_{i+j} = \int_{c_l}^{c_u} dx g(x) \exp[iS(x)] \quad (2.130)$$

where $S(x)$ is the action, and x_i, x_j represent the two saddle points, in our case the short and long. The following change of variable permits to have both saddle points in the same equation. Writing the action in a cubic order

$$S(x) = \bar{S} + \epsilon x - ax^3, \quad g(x) = g_0 + g_1 x \quad (2.131)$$

where the two saddle points obtained from $S'(x) = 0$ are

$$x_{i,j} = \pm \sqrt{\frac{\epsilon}{3a}} \quad (2.132)$$

both saddle points coalesce when $\epsilon = 0$. $\bar{S}$ is the value of the action in the middle point between both saddle points. The decomposition of $g(x)$ in a linear function permits to use this approximation near the cutoff $g(x) = g_0$, and away from it. In this way, the numerical solution is the same for all saddle points regardless of where they are placed, near or far from the cutoff point.

The solution of the integral from Eq. (2.130) is given by

$$\begin{aligned} M_{i+j} &= \sqrt{\frac{2\pi\Delta S}{3}} e^{(i\bar{S}+i\pi/4)} \\ &\times \{ \Delta A [J_{1/3}(\Delta S) + J_{-1/3}(\Delta S)] + \bar{A} [J_{2/3}(\Delta S) - J_{-2/3}(\Delta S)] \} \quad (2.133) \\ \Delta S &= (S_i - S_j)/2, \quad \bar{S} = (S_i + S_j)/2 \\ \Delta A &= (A_i - A_j)/2, \quad \bar{A} = (iA_i - A_j)/2 \end{aligned}$$

where

$$\begin{aligned} \Delta S &= 2\epsilon^{3/2}(27a)^{-1/2} \\ \bar{A} &= g_0(-2\pi i)^{1/2}a^{1/4}(3\epsilon)^{-1/4} \\ \Delta A &= g_1(2\pi i)^{1/2}\epsilon^{1/4}(3a)^{-3/4} \end{aligned} \quad (2.134)$$

Figure 2.5: Figure shows a comparison of spectra using the saddle point approximation with and without the uniform approximation. The black line corresponds to a spectrum calculated using the uniform approximation. Red line represents an spectrum calculated without this approximation. The spectra has been generated by a laser with a central wavelength of $\lambda = 800 \text{ nm}$, and with an intensity of $3.0 \times 10^{14} \text{ W/cm}^2$ for the neon atom using the dipolar transition matrix given in Eq. (2.106) Fig. (a) Full spectrum, Fig. (b) this spectrum corresponds to a single cycle of the laser.

Later on Milosevik and Becker given a simplified mathematical form based on the Airy functions [71], [86]:

$$\begin{aligned}
 M_{i+j} &= M_s \exp(iS_i) + M_l \exp(iS_j) \\
 &= (6\pi S_-)^{1/2} \exp(iS_+ + i\pi/4) \\
 &\times \left[\frac{A_-}{\sqrt{z}} \text{Ai}(-z) + i \frac{A_+}{z} \text{Ai}'(-z) \right] \phi(\omega)
 \end{aligned} \tag{2.135}$$

where $\text{Ai}(z)$, $\text{Ai}'(z)$ are the Airy function and its derivative, being z a complex

variable.

$$\begin{aligned}
z &= \left(\frac{3S_-}{2} \right)^{2/3} \\
2S_{\pm} &= S_i \pm S_j \\
2A_{\pm} &= A_i \pm iA_j
\end{aligned} \tag{2.136}$$

where S_i and S_j are the values of the action at the saddle points x_i, x_j . The value of z must be replaced by $z \exp(-2i\pi/3)$ for the harmonics $\omega > \omega_c$. In addition, to have a right phase is necessary to add a new term $\phi(\omega)$ although, it was not reported on any paper perhaps because it was not tested. That is easy to check comparing the phase of the dipole modulus for the cases with and without the uniform approximation.

$$\begin{aligned}
\phi(\omega) &= 1, \quad \omega < \omega_c \\
\phi(\omega) &= \exp(-i\pi), \quad \omega > \omega_c
\end{aligned} \tag{2.137}$$

Fig. 2.5(a) exhibits a comparison of the spectra with and without the uniform approximation. Practically both spectra have the same values. Fig. 2.5(b) shows the spectra produced by a single cycle, here the differences are near the cutoff points. Fig. 2.5(b) shows a jump around the 55th harmonic. This jump is the result of removing all points of the frequencies higher than the point where the trajectory start to diverging. On the contrary, the uniform approximation exhibits a soft transition and the jump is removed. For the region far from the cutoff both curves present same values.

2.4.7 Classical trajectories.

Deeper insight in the physical processes could be obtained regarding the movement of the electrons when are ionized. [71] [86] [51]. The electrons follow in their movements the Newton's classical equations. Then, the position of the electron is determined by the saddle points $(\mathbf{k}_{qs}, \tau_{qi}, \tau_{qr})$

$$\mathbf{r}(t) = \mathbf{k}_{qs}(t - \tau_{qi}) + \int_{\tau_{qi}}^t \mathbf{A}(t') dt' \tag{2.138}$$

where the position at the ionization time is at the origin $\mathbf{r}(\tau_{qi}) = 0$ and the same point for the recombination time $\mathbf{r}(\tau_{qr}) = 0$. The position $\mathbf{r}(t)$ is a complex magnitude.

For times greater than the ionization time, t must be real, until the moment of recombination where $t = \tau_{qr}$ is complex again. Then $\Re[\tau_{qi}] < t < \Re[\tau_{qr}]$. This expression allows knowing what is the trajectory followed by an electron that emits a photon with the ω frequency.

The distance from the parent ion is given by

$$d(t) = (\Re[r_x(t)]^2 + \Re[r_y(t)]^2)^{1/2} \quad (2.139)$$

However, how $d(\tau_{qi}) \neq 0$ and $d(\tau_{qr}) \neq 0$ is interpreted as the electron never starts or ends to the parent ion. The value $d(\tau_{qi})$ is interpreted as the distance at the electron enters in the continuum after the tunneling ionization. This distance is the order of few atomic units.

Figure 2.6: Figure shows how an electron moves away over time before it recombines and emits a photon of a specific frequency. The red line corresponds to the short trajectory, the blue line corresponds to the long trajectory. The black lines indicate that trajectories short and long are the indistinguishable. The electric field is plotted in the background. Fig. (a) corresponds to the distances when the photon emitted is the 49th harmonic. Fig. (b) is for the 35th harmonic.

Figs. 2.6(a)(b) shows the electron is freed at the distance of 10 *au*. independently of the frequency of the photon emitted at the recombination time. Both Figs. 2.1 and 2.6 correspond to the same experiment. The black lines in Fig.

2.6(a) indicates that the electron follows the same paths independently whether they are short or long. This can be confirmed in Fig. 2.1(b) where the 49th harmonic is in the cutoff region of the spectrum for the second and third visit to parent ion until the recombination time. On the contrary, the 35th harmonic is in the plateau region of the spectrum where all trajectories are clearly separated each other.

Another magnitude can be determined from the saddle points is the velocity that is calculated of the expression $\mathbf{v}(\mathbf{k}, t) = \mathbf{k} + \mathbf{A}(t)$ then

$$\mathbf{v}(t) = \mathbf{A}(t) - \mathbf{A}(\tau_{qi}) \quad (2.140)$$

where k_{qs} is a constant along the trajectory followed by the electron. Also, the initial velocity of the electron at ionization time is zero. Finally, the kinetic energy is calculated from the velocity, that in the atomic units is

$$E_{kin}(t) = \frac{[\mathbf{A}(t) - \mathbf{A}(\tau_{qi})]^2}{2} \quad (2.141)$$

A deeper study of the photoelectrons trajectory must be included the atomic Coulomb potential. A detailed description about how to implement numerically this procedure is in paper and thesis from Yan Tina-Min [102], [101]

2.4.8 Ionization.

When the intensity of the laser is high enough there is a significant probability that the atom is ionized over time. Then, when the ground state is depleted, the photon emission is suppressed. The methodology followed by the time-evolution operators does not treat the ground state depletion; despite the original Lewenstein model (2.31) is covered by the factor $a(t)$. To fill this gap, the evolution of the amplitude in the ground state is included in Eq. (2.92) as the following multiplicative coefficient

$$\Gamma(t) = \exp\left(-\int_{t_0}^t W[\mathbf{E}(t')]dt'\right) \quad (2.142)$$

where $W[\mathbf{E}(t')]$ is the ionization rate, $\Gamma(t)$ is the probability the electron remains bound. So, $1 - \Gamma(t)$ indicates the ionization probability.

An analytical expression derived from the tunneling ionization rate of the complex atoms in an AC electric field is the Ammosov, Delone, and Krainov equation [3]. The initial state is described by an effective quantum number n^*

and the angular and magnetic quantum numbers by l and m respectively. The ADK equation is as following

$$W_{ADK} = C_{n^*l}^2 f(l, m) \frac{\kappa^2}{2} \left(\frac{3E}{\pi\kappa^3} \right)^{1/2} \left(\frac{2\kappa^3}{E} \right)^{2n^* - |m| - 1} \exp \left(-\frac{2\kappa^3}{3E} \right). \quad (2.143)$$

with

$$C_{n^*l} = \left(\frac{2}{n^*} \right)^{n^*} (2\pi n^*)^{-1/2} \quad (2.144)$$

$$f(l, m) = \frac{(2l+1)(l+|m|)!}{2^{|m|}|m|!(l-|m|)!}$$

Eq. (2.143) is obtained averaging over a single laser cycle. Although above formula is right, the simplified expression with $m = 0$ and $l = 0$ in the original paper contains an error [29], the π/e factor is missing. The validity of the formula is for $n^* \gg 1$, the frequency of the laser field is much lower than the ionization energy $\omega \ll I_p$, and the intensity of the laser is not so high to suppress the potential barrier.

It is known that the ADK equation could give an overestimation of the ionization rate when is applied to intense and short pulses [67]. Other expressions to calculate the ionization are in paper of Bauer [16] and Tong. [97]

2.5 TDSE: Qprop program.

The code Qprop was developed to study the laser-atom interactions in a non-perturbative and non-relativistic regimen that is where the high-order harmonics generation appears. The single active electron (SAE) approximation is solved by the TDSE in three spatial dimensions. The study of the multielectron atoms is solved by the Kohn-Sham (KS) equations. In both cases, the wavefunctions are expanded by the spherical harmonics. Time propagation of the wavefunctions is performed using a split-operator approach.

2.5.1 Time-dependent density functional theory

Density functional theory (DFT) was introduced by Hohenberg and Khon [47] [77]. DFT is an alternative variational procedure to the solution of the Schrödinger equation, where the functional of the electronic energy is minimized with respect to the electronic density $n(\mathbf{r})$.

The basic quantity of the density-functional theory is the electron density $n(\mathbf{r})$, that is always a function of three space coordinates and time and independent of the number of particles. In practice, the time-dependent density functional theory is formulated via the Kohn-Sham equation

$$i\frac{\partial}{\partial t}\Psi_i(\mathbf{r}, t) = H_{KS}\Psi_i(\mathbf{r}, t), \quad i = 1, 2, \dots, N \quad (2.145)$$

where $\Psi_i(\mathbf{r}, t)$ are the Kohn-Sham orbitals, and H_{KS} is the Kohn-Sham Hamiltonian. $\Psi_i(\mathbf{r}, t)$ are not wavefunctions in the sense of the Schrödinger equations; its physical significance is to build the electron density according to the following equation

$$n(\mathbf{r}, t) = \sum_{i=1}^N |\Psi_i(\mathbf{r}, t)|^2 \quad (2.146)$$

The Kohn-Sham Hamiltonian H_{KS} and Schrodinger Hamiltonian H differs only by the effective potential $\phi_{KS}(\mathbf{r}, t)$

$$H_{KS} = \frac{1}{2} [\mathbf{p} + \mathbf{A}(t)]^2 + V(\mathbf{r}) + V_{ee}[n(\mathbf{r}, t)] \quad (2.147)$$

where $V(\mathbf{r})$ is the ionic potential, and $V_{ee}[n(\mathbf{r}, t)]$ is the electron-electron interaction potential that comprises the Hartree-part $U[n(\mathbf{r}, t)]$, and the exchange-correlation potential $V_{xc}[n(\mathbf{r}, t)]$.

$$V_{ee}[n(\mathbf{r}, t)] = U[n(\mathbf{r}, t)] + V_{xc}[n(\mathbf{r}, t)] \quad (2.148)$$

where the Hartree potential is defined as

$$U[n(\mathbf{r}, t)] = \int \frac{n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}' \quad (2.149)$$

In practice, the exchange correlation potential $V_{xc}[n(\mathbf{r}, t)]$ has to be approximated.

The interaction potential for the ground state and the central field approximation $V_{ee}[n(\mathbf{r}, t)] \rightarrow V_{ee}[n(r, t)]$. Instead under the interaction of a laser linearly polarized

$$V_{ee}[n(\mathbf{r}, t)] = V_{ee}^0(r, t) + V_{ee}^1(r, t)\cos\theta + V_{ee}^2(r, t)\frac{1}{2}(\cos^2\theta - 1) \quad (2.150)$$

The KS orbitals are expanded in spherical harmonic functions

$$\Psi_i(\mathbf{r}, t) = \frac{1}{r} \sum_{l=0}^{\infty} \sum_{m=-l}^l \Phi_{ilm}(r, t) Y_l^m(\theta, \phi) \approx \frac{1}{r} \sum_{l=0}^L \sum_{m=-l}^l \Phi_{ilm}(r, t) Y_l^m(\theta, \phi) \quad (2.151)$$

When the laser is linearly polarized, the quantum number m remains unaltered

$$\Psi_i(\mathbf{r}, t) = \frac{1}{r} \sum_{l=0}^{\infty} \Phi_{ilm_i}(r, t) Y_l^{m_i}(\theta, \phi) \approx \frac{1}{r} \sum_{l=0}^L \Phi_{ilm_i}(r, t) Y_l^{m_i}(\theta, \phi) \quad (2.152)$$

where L is the maximum value can reach the quantum number l . The number L is one of the parameters of the grid and should be large enough to avoid lost of data. This value depends on the pulse length and the laser intensity. For the unperturbed atom the so-called central field approximation (CFA) is usually adopted. This leads in the ground state KS orbitals of the form

$$\Psi_i(\mathbf{r}, t) = \frac{\Phi_{ilm_i}(r, t)}{r} Y_l^{m_i}(\theta, \phi) \quad (2.153)$$

as a consequence of the interaction with the laser the radial part Φ_{ilm_i} changes its distribution along r also due to the coupling of the field is populated the Φ_{il-1m_i} and Φ_{il+1m_i} . The evolution of the radial part is given by

$$\begin{aligned} i \frac{\partial}{\partial t} \Phi_{ilm_i}(r, t) = & \left(-\frac{1}{2} \frac{\partial^2}{\partial r^2} + V(r) + \frac{l(l+1)}{2r^2} \right) \Phi_{ilm_i}(r, t) \\ & - iA(t)r \sum_{l'} \langle Y_l^{m_i} | \cos\theta | Y_{l'}^{m_i} \rangle \frac{\partial}{\partial r} \frac{1}{r} \Phi_{il'm_i}(r, t) \\ & + iA(t) \sum_{l'} \langle Y_l^{m_i} | \sin\theta \frac{\partial}{\partial \theta} | Y_{l'}^{m_i} \rangle \frac{1}{r} \Phi_{il'm_i}(r, t) \\ & + \sum_{l'} \langle Y_l^{m_i} | V_{ee}[n(\mathbf{r}, t)] | Y_{l'}^{m_i} \rangle \Phi_{il'm_i}(r, t) \end{aligned} \quad (2.154)$$

When Qprop uses the single electron approximation, the interaction potential electron-electron $V_{ee}[n]$ is not used, then last term in above equation is removed and the Kohn-Sham equation becomes the Schrödinger equation. The above equation can be written as

$$i \frac{\partial}{\partial t} \Phi(\mathbf{r}, t) = (\mathbf{H}_{int}(t) + \mathbf{H}_{at}) \Phi(\mathbf{r}, t) \quad (2.155)$$

where $\Phi(\mathbf{r}, t)$ is a vector which components are in the space l where $l = 0, 1, \dots, L$. The Hamiltonian are splitted in one part time dependent and another time independent. To calculate the evolution of the function $\Phi(\mathbf{r}, t)$ is used the time-evolution operator.

Figure 2.7: Spectrum of the neon. Laser intensity $5.0 \times 10^{14} \text{ W/cm}^2$ $\lambda = 1053 \text{ nm}$. The DFT curve has been obtained using the DFT model from Qprop using all electrons of the neon atom. The SAE curve was obtained from Qprop using a single electron.

2.5.2 Discrete time propagators

The temporal evolution of any state can be expressed in function of the time-evolution operator as

$$|\Psi(t)\rangle = U(t, t_0)|\Psi(t_0)\rangle \quad (2.33)$$

A numerical approximation of the time-evolution operator is given by

$$U(t_{i+1}, t_i) = \exp[-i\Delta t H(t_{i+1/2})] \quad (2.156)$$

where has been used the approximation of the Hamiltonian varies smoothly over time. For small time steps, a good approximation is to take the central value in the time step for the Hamiltonian.

$$U(t_{i+1}, t_i) = I - i\Delta t H(t_{i+1/2}) \quad (2.157)$$

Unfortunately, this operator does not preserve the norm of the wavefunction because it is not an unitary operator (2.37).

The evolution propagator used to solve Qprop and others TDSE solvers is based on the scheme Peaceman-Rachford that is defined as

$$\begin{aligned} U(t_{i+1}, t_i) = & \left[I + i\frac{\Delta t}{2} H_{int}(t_{i+1}) \right]^{-1} \left[I + i\frac{\Delta t}{2} H_{at} \right]^{-1} \\ & \times \left[I - i\frac{\Delta t}{2} H_{at} \right] \left[I - i\frac{\Delta t}{2} H(t_i) \right] \end{aligned} \quad (2.158)$$

A detailed implementation of this algorithm can be found in the Muller paper [74].

2.5.3 Imaginary time propagation.

Before the interaction with a laser the atom is in the ground state (the state with minimal energy) and from here, the atom evolves under the interaction laser-atom. Qprop uses the previous propagator algorithm to determine the ground state replacing $\Delta t \rightarrow i\Delta t$. The evolution operator for an unperturbed atom is $U(t, t_0) = \exp(-iE_n\Delta t)$. When the following equation

$$|\Psi(t)\rangle = U(t, t_0)|\Psi(t_0)\rangle \quad (2.33)$$

is solved iteratively, the value of $\Psi_n(\mathbf{r}, t)$ is reduced in each iteration because the time is complex. This reduction is stronger when the energy level is higher. After each iteration $\Psi_n(\mathbf{r}, t)$ is renormalized, then only the wavefunction with lower energy remains after several iterations. The same procedure is applied for boht SAE or TDDFT; although for the TDDFT the time to converge in the ground state increases with the number of electrons.

2.5.4 Imaginary absorbing potential.

An undesired effect appears when the density of charge from the electrons approach to the boundaries limits is the reflection of the density occasioning a misunderstanding results. Qprop has an absorption algorithm near boundaries implemented by the imaginary potential $iV_{im}(r)$. This potential is added the Schrödinger equation. This function is defined to be zero in the area around the atom and increases progressively up to the edge of the grid. The procedure to vanish the wavefunction is the same as before section, an imaginary potential is an attenuation in the time-evolution operator.

2.5.5 The gauge.

Although from the analytical point of view the selection of the velocity or position gauge should be produce similar results, when the TDSE is solved numerically the results are different. The selection of the wrong gauge can ruin completely the simplification of the spherical description introducing a increasing detail in the angular dependency. The value of the interaction Hamiltonian in the length gauge $\mathbf{E}(t) \cdot \mathbf{r}$, increases with the distance and them the far way the region of the wavefunction is more weighted than the nearest part to the nucleus where the norm of the wavefunction has the greater values.

Figure 2.8: Spatial distribution of the neon wavefunction for a linearly polarized laser obtained with Qprop. The intensity is $2.0 \times 10^{14} \text{ W/cm}^2$ $\lambda = 800 \text{ nm}$. The double bubble in the center of the figure indicates is the $2p$ orbital of the neon.

The general expression of the derivative of a magnitude, in atomic units

$$\frac{d\xi}{dt} = i[H\xi] + \frac{\partial \xi}{\partial t} \quad (2.159)$$

where the Hamiltonian adopts one of two forms in length or velocity gauge (2.16) (2.10). The expression of the velocity changes according the gauge. From Eq. (2.159) changing $\xi \rightarrow x$, in the length gauge $\mathbf{v} = \mathbf{p}$ and in the velocity gauge $\mathbf{v} = \mathbf{p} + \mathbf{A}(t)$ where $\mathbf{p}$ is a constant of motion.

A laser with $E_0 = 0.05 \text{ au} \approx 1 \times 10^{14} \text{ W/cm}^2$ and $\omega = 0.05 \text{ au} \approx 800 \text{ nm}$ has for the vector potential $A(t) = 1 \text{ au}$.

In the velocity gauge, once the electron escapes from the atomic potential, it quivers as $r = pt + (E_0/\omega^2)\sin \omega t$, with $r \approx 20 \text{ au}$, $p = 1 \text{ au}$ and thus, the angular momentum is $l = r \times p \approx 20$. In the length gauge $v \approx A(t)$, the maximum value is $v \approx 1$ so, the value for $l = r \times p$ is proportional to r . The maximum value of r depends on the type of problem. For ionization processes

a radius with $r = 50 \text{ au}$ is big enough, on the contrary, for high-order harmonics generation r could be in a range between $300 - 1000 \text{ au}$.

The comparison between the length and velocity gauges shows that in the velocity gauge the size of the mesh L in Eqs. (2.152) (2.151) is much smaller than in the length gauge for a similar result. [74].

2.6 Tools.

2.6.1 Time-frequency analysis.

One of the great advantages of the saddle point approximation is to know for a single harmonic some important magnitudes such as the intensity, the recombination, and ionization time. In this way, is possible to see how every trajectory short and long contributes to the spectrum. On the contrary, when a Schrödinger solver (TDSE) like Qprop [13] or the Lewenstein model from Eq. (2.95) is used the information from the long and short trajectories is hidden. This information can be recovered by the time-frequency analysis (TFA) [30]. The TFA uses the spectrum obtained from the Fourier transform of the dipole moment together with a window that is shifted from lowest to highest frequencies. When the window is narrower, the frequency resolution is higher, in contrast, the time resolution is poor. The size of the window is fixed manually by means of the parameter σ .

$$d_{TFA}(\omega, t) = \int d(t') \exp(-i\omega t') \frac{\exp[-(t' - t)^2/2\sigma^2]}{\sigma\sqrt{2\pi}} dt' \quad (2.160)$$

where the last term of the integral is a Gaussian function that is used as window, and $d(t')$ is the temporal evolution of the dipole moment. The size of windows is adjusted to $\sigma = 1/(3\omega)$ where ω is the central frequency of the laser. In addition, TFA could be used as a roughly validation of results obtained with the saddle point approximation.

Fig. 2.9(a) corresponds to a laser pulse with orthogonal polarization whose frequencies are $\omega, 2\omega$, and Fig. 2.9(b) corresponds to a linearly polarized laser. For Fig. 2.9(a), the weight of the short trajectories of the dipole moment are greater than the long ones. It can be interpreted as the electrons that follow the long trajectories practically never return. On the other hand, when the electron does not recombine at the first visit to the parent ion, the subsequent recombinations can be neglected. This is because there is not any kind of structure

under the bells (the pink-lines) as happen in Fig. 2.9(a). This structure indicates the presence of the recombinations corresponding to the second or third visit to parent ion.

Fig. 2.9(b) both short and long paths contribute in a similar way to the dipole modulus. Under the bells there are several lines that suggest the recombination of the electron in the second or third visit to the atom.

The spectra from both figures can be obtained from frequencies greater than the 40th harmonic, without loss of precision, using only the short and long trajectory from the first visit to the parent ion.

2.6.2 The inverse Fourier transform.

One of the most interesting applications is the inverse Fourier transform when is applied to a range of the spectrum. When we have a spectrum of a polarized orthogonal pulse, this tool allows to observe the elliptic shape of the harmonic, the angle of rotation of the ellipse and the magnitude of its X and Y components . Therefore, when this harmonic is used as a probe for circular dichroism, we know how the electric field is before and after interacting with the matter to subsequently analyze the effects of interaction.

For a linearly polarized laser, the reconstructed temporal signal is a set of periodically spaced peaks. Some control about the wide of the peaks is possible by selecting the range of harmonics. The peaks obtained by this procedure are the most shorted electric field pulses ever produced. They can reach the attosecond length or even zeptoseconds using a laser with wavelength around 4000 nm [45]. Schafer and Gaarde [88] showed that the use of two electric fields with different frequencies can modify the quantum path to define high resolution attosecond clock.

Fig. 2.10 exhibits a temporal pulse generated by the dipole moment from the neon for two cases: Fig. 2.10(b) is the temporal evolution from a single electron 2p with quantum number $m = -1$. Fig. 2.10(a) is the simultaneous contribution from both electrons $m = \pm 1$. In both figures the field produced is LCP (Left Circularly Polarized) along the length of the pulse. Fig. 2.10(a) shows the contribution of both electrons duplicates the size of the pulse with light-blue color from Fig. 2.10(b), see the projected image. For the rest of sections of the pulse perhaps there is a destructive interference that reduces the intensity.

Figure 2.9: TFA spectrum from neon. Spectra generated with a laser with central wavelength of $\lambda = 800 \text{ nm}$. Fig. (a) corresponds to the X component of two collinear lasers where the field $E_x(t)$ is described by $\lambda = 800 \text{ nm}$, $I = 4.0 \times 10^{14} \text{ W/cm}^2$ envelope $\sin^2$, length 7 cycles and $E_y(t)$ is $\lambda = 400 \text{ nm}$, $I = 1.0 \times 10^{14} \text{ W/cm}^2$, the envelope used is $\sin^2$ to simulate a Gaussian function the length of the pulse is 14 cycles. Fig. (b) is a linearly polarized laser whose field coincides with the E_x of Fig. (a). The pink-lines are the calculation with the saddle approximation using only the first visit to the parent ion.

Figure 2.10: Temporal evolution of pulse gets from the inverse Fourier transform. The spectrum window selected to calculate the inverse Fourier transform is the range of $33^{th} - 80^{th}$ harmonics. The laser pulse is orthogonally polarized: the X-axis $\lambda = 800\text{nm}$, $I = 4.0 \times 10^{14}\text{W/cm}^2$ with a $\sin^2$ envelope formed by 7 cycles, and the Y-axis $\lambda = 400\text{nm}$, $I = 1.0 \times 10^{14}\text{W/cm}^2$ with a $\sin^2$ envelope formed by 14 cycles. The atom is neon. Fig. (b) shows the temporal evolution generated from a single electron from the $2p$ orbital with the quantum number $m = -1$. Fig. (a) shows the addition of the contribution of both electrons $m = \pm 1$ from the $2p$ orbital.

Chapter 3

Results: Mixture of gases helium-neon

3.1 Introduction

This chapter is a theoretical study about the interference of the harmonics generated by a mixture of two gases helium-neon. The model is based on the electron quantum paths, and the bound-free transitions. The interaction of a laser with intensity around $10^{14}W/cm^2$ with a mixture of gases, helium-neon, produces an interference that is destructive at the low order harmonics and oscillates between constructive and destructive near cut-off. This destructive interference at high harmonic may be used to explore other transitions which are currently hidden behind the strong signal of current processes. Our numerical results are in very good agreement with experimental data in the range of the low harmonics frequency, At higher-order harmonics, where there are not experimental data, the comparison is with the Schrödinger solver (Qprop).

This chapter is organized as follows. The theoretical section contains the mathematical expressions of the dipole moment as the sum of the quantum paths. By means of the Fourier transform over the dipole moment one get the phase of each harmonic. In the following, we show that the phase of each harmonic depends on the matrix expression of dipolar transition matrix, is to say, the expression of the wavefunction of the electron on the ground state. We test different models of dipolar matrix based on the Gaussian wavefunction [64], Slater-type orbitals (STO) [44], asymptotic wavefunction [41], and select the one that best fit to the solution of the Schrödinger solver [14] for high-order

harmonics. Then, based on the relative phase for each gas helium-neon and the rates of its density we obtain a interference pattern per each harmonic. Here, the parity of the outermost electron of the neon is critical. Most of the models based on hydrogenic atoms reproduces correctly the ground state for the $1s$ orbital with even parity. On the contrary for the neon, the most external electron in the ground state is $2p$ orbital with odd parity, then to use a model based on $1s$ orbitals has a consequence a wrong phase selection. [15] How this chapter study the interference between harmonics, the correct phase selection is decisive for the calculations.

The numerical results section is dedicated to the study of the relative phase of the harmonics of a mix of two noble, gases helium-neon, under the interaction of a laser linearly polarized with an intensity around $4 \times 10^{14} \text{W/cm}^2$. This section is divided in several parts. First, we compare the simulations of the relative phase between the gases, based on the quantum paths, with the experimental data of Kanai [53]. How the experimental data only covers the low and mid-range of the harmonics spectrum and then the highest-order harmonics are compared with other simulations based on the Schrödinger solver.

Second, the spectrum of the harmonics is decomposed in a sum of the quantum paths from which is select those paths with the higher values. As a result, we select the short and long quantum paths that represent the electron trajectories which recombines in the first visit to the parent ion. This simplification allows us to explain the experimental results at the lower-order harmonics and the oscillations that appear at the high-order harmonics in the spectrum. How the ionization can be depreciated for the intensities used in the calculations, therefore the sum of the quantum paths allows us to extrapolate the results to lasers with longer pulses.

Third, based on the recombination and ionization times, we can see that the relative phase does not change appreciably when the intensity of the laser is increased. Independently of intensity of the laser, the relative phase is similar at the lower-order harmonics and the oscillations remain at the harmonics near cut-off. In the middle range of the harmonics spectrum, where there is not a clear predominant quantum path, it is a little risky to extract some conclusions from the calculations.

Finally, the change in the ratio between the densities of the gases changes the intensity of the harmonics; some of them have constructive interference and others destructive. This could be used as some kind of equalization of the intensity of harmonics and in lesser extend the phase of the harmonic. The

Figure 3.1: Phase of neon harmonics calculated from the Fourier transform of the expression in Eq. (2.95) with an intensity $I = 4 \times 10^{14} \text{ W/cm}^2$. Comparison using different dipole transitions matrix $\langle k|r|\Psi_0 \rangle$: STO is based on the Slater Type Orbitals for neon (3.3), GAUS is using a dipolar transition matrix based on Gaussian wavefunction and Qprop is an Schrödinger solver.

destructive interference might show other atomic processes currently hidden by the intensity of bound-free transitions.

3.2 Theoretical Model

First, we consider an atom in a single-electron approximation under the influence of an electric field with linear polarization. The Lewenstein expression [64] using the length gauge $H_{int}(t) = E(t)r$ is given by

$$d(t_r) = i \int_{t_0}^{t_r} dt_i \int dk \langle \Psi_0 | r | v(k, t_r) \rangle \langle v(k, t_i) | E(t_i) r | \Psi_0 \rangle e^{-iS(k, t_i, t_r)} + cc \quad (2.95)$$

where the dipolar transition matrix is the unknown term, and it was calculated in the section (2.4.4) and section (2.4.5)

How the dipolar transition matrix depends on the form of the wavefunction, we compare three cases: the Gaussian [64], the Slater Type Orbital [44] and the last one based on the asymptotic behavior of the wavefunction [41]. In the following calculations, the dipolar transition matrix based on the Slater Type Orbitals is used because it produces a phase similar to that of the Schrödinger solver (Qprop) at higher-order harmonics.

All dipolar transition matrix based on the orbitals $1s$ reproduces wrongly the phase for neon, as a consequence the difference of the phase between helium-neon has an error of $\pi/2$ such as happened with the Gaussian model. See higher harmonics on Fig. 3.1. That say, the expression of the dipolar transition matrix in Eq. (3.3) produces different result for the neon when it compares with the helium. When the velocity recombination change from $k \rightarrow -k$, in the helium $\langle -k|r|\Psi_0 \rangle_{He} = -\langle k|r|\Psi_0 \rangle_{He}$ on the contrary for the neon $\langle -k|r|\Psi_0 \rangle_{Ne} = \langle k|r|\Psi_0 \rangle_{Ne}$ [15] However, if our intention is to study the dipole modulus, the asymptotic model produces better result compared with Qprop.

The wavefunction of the Slater orbitals in the momentum space [44]:

$$\langle k|\Psi_0 \rangle = F_{nl}(k) Y_{lm}(\theta, \phi) \quad (3.1)$$

where $F_{nl}(k)$ is given by Eq. (2.104). For the particular cases of the helium and neon with linear polarization, we have

$$\begin{aligned} \langle k|\Psi_0 \rangle_{He} &= \frac{2^{15/4} I_p^{5/4}}{\sqrt{\pi}(k^2 + 2I_p)^2} \frac{1}{\sqrt{4\pi}} \\ \langle k|\Psi_0 \rangle_{Ne} &= \frac{2^{25/4} I_p^{7/4} k}{\sqrt{3\pi}(k^2 + 2I_p)^3} \frac{\sqrt{3}}{\sqrt{4\pi}} \end{aligned} \quad (3.2)$$

Using the relationship between the wavefunctions in the momentum space, then the transition matrices are given by

$$\langle k|r|\Psi_0 \rangle = i \frac{d}{dk} \langle k|\Psi_0 \rangle \quad (2.98)$$

And finally, the dipolar transitions matrices are

$$\begin{aligned} \langle k|r|\Psi_0 \rangle_{He} &= -i \frac{2^{23/4} I_p^{5/4} k}{\sqrt{\pi}(k^2 + 2I_p)^3} \frac{1}{\sqrt{4\pi}} \\ \langle k|r|\Psi_0 \rangle_{Ne} &= -i \frac{2^{25/4} I_p^{7/4} (5k^2 - 2I_p)}{\sqrt{3\pi}(k^2 + 2I_p)^4} \frac{\sqrt{3}}{\sqrt{4\pi}} \end{aligned} \quad (3.3)$$

Calculating the Fourier transform in Eq. (2.95) by means of the saddle point approximation together with the integration by parts to remove the electric field in the final equation, the result gives a superposition of the quantum paths given by Eq. (2.123). Where $M(v)$ and ν in Eq. (2.124) for the helium and neon are given by

$$\begin{aligned} M_{He}(v) &= \frac{2^{15/4} I_p^{5/4}}{\sqrt{\pi}} \frac{1}{\sqrt{4\pi}}, \quad \nu = 2 \\ M_{Ne}(v) &= \frac{2^{25/4} I_p^{7/4} k}{\sqrt{3\pi}} \frac{\sqrt{3}}{\sqrt{4\pi}}, \quad \nu = 3 \end{aligned} \quad (3.4)$$

By the way, if the quantum paths are decomposed as module-argument format

$$d(\omega) = \sum_q d_{(q)}(\omega) = \sum_q |d_{(q)}(\omega)| e^{i\phi_q(\omega)} \quad (3.5)$$

as a result, this summation can be seen as an interference between several trajectories for the same harmonic ω . According to the recombination times in the first, second, and third visit to the parent ion until they recombine, we have:

$$\begin{aligned} d(\omega) = & \sum_q d_{l(q)}^1(\omega) + d_{s(q)}^1(\omega) \\ & + \sum_q d_{l(q)}^2(\omega) + d_{s(q)}^2(\omega) + \sum_q d_{l(q)}^3(\omega) + d_{s(q)}^3(\omega) \end{aligned} \quad (3.6)$$

where the upper index of the dipole values indicates when the electron recombines with the parent ion: Index 1 is when $0 < \Re[\tau_r - \tau_i] < T$ electron recombines in the first visit to the parent ion, index 2 is when $T < \Re[\tau_r - \tau_i] < 2T$ recombination in the second visit to the parent ion and index 3 is when $2T < \Re[\tau_r - \tau_i] < 3T$ for the third visit to the parent ion. The bottom Fig. 3.5 has the dipole modulus for the short and long trajectories with different recombination times.

The total intensity generated by two electric fields as a result of its interference has the expression $|E_1(\omega) + E_2(\omega)|^2 = |E_1(\omega)|^2 + |E_2(\omega)|^2 + 2\Re[E_1(\omega)E_2^*(\omega)]$. Based on that, the intensity for a particular harmonic ω [53] depends on the intensity of the dipole value $d(\omega)$ in Eq. (2.95) and the density ρ of gas, as

$$I(\omega) = \rho_1^2 |d_1(\omega)|^2 \left| \left(1 + \frac{r_2}{1 - r_2} \frac{d_2(\omega)}{d_1(\omega)} e^{i\Delta\phi(\omega)} \right) \right|^2 \quad (3.7)$$

where

$$\Delta\phi(\omega) \equiv \phi_2(\omega) - \phi_1(\omega) \quad (3.8)$$

is the relative phase of the harmonics from each gases 1 and 2. Only the atoms in the ground state contribute significantly to the atomic dipole value.

The condition for constructive interference is given by $\Delta\phi(\omega) = 2\pi n$ ($n \in \mathbb{Z}$) and destructive interference $\Delta\phi(\omega) = (2n + 1)\pi$, where ρ_i is the density of the gas i , and r_i is the rate of the gas i $r_i = \rho_i / \rho_{total}$ with $\rho_{total} = \rho_1 + \rho_2$. Therefore, we can modify the intensity for a particular harmonic ω tuning the

rate r_i and $\Delta\phi(\omega)$. Only for the harmonics with constructive phase we have an improvement for the intensity of this harmonic. And for harmonics with destructive phase $\Delta\phi = \pi$, a proper ratio between both gases are needed to have a total destructive interference.

If we treat the interference as an addition of two complex numbers where the phase comes from the atomic dipole phase for every gas and the module for every gas comes from the total contribution from all atoms, we can adjust the rate between the complex modules changing the ratio density, in this way, we can modify the phase of the electromagnetic field of the harmonic. Following section, we can see how much $\Delta\phi(\omega)$ changes for a combination of two gases helium-neon.

To appreciate the interference of the harmonics using two gases, the macroscopic dipole moment $D_i(t) = \rho_i(t)d_i(t)$ should have a similar value for both gases. Due to $d_i(t)$ is an intrinsic value of the atom, only is possible to modify $D_i(t)$ modifying the density of gas $\rho_i(t)$.

When we include the ionization in our calculations, the phase of the dipole moment remains unchanged. The ionization only modifies the dipole modulus because it is real magnitude, then the phase is not modified. By the way, the ionization modifies the density of the atoms in the ground state, therefore, the ratio between the density of the gases in the ground state is changed and the interference constructive or destructive might be not true at the time.

Figure 3.2: Fig. (a) The continues black line is the spectrum obtained with a mix of the gases where number of helium atoms is 33 times higher than neon for the intensity $4.0 \times 10^{14} W/cm^2$. The 61th harmonic appears practically annihilated such as it was calculated in Fig. 3.7, Fig. (b): constructive interference for the 59th harmonic

Figure 3.3: Relative phase $\Delta\phi = \phi_{He} - \phi_{Ne}$ with a intensity of the laser of $I = 4.0 \times 10^{14} \text{ W/cm}^2$ and $\lambda = 800 \text{ nm}$. (QP) Relative phase calculated by means quantum paths. Exp. Experimental from Kanai. [53].

3.3 Numerical Results

The laser used for calculations is linearly polarized with a wave length of $\lambda = 800 \text{ nm}$ and an intensity of $4.0 \times 10^{14} \text{ W/cm}^2$. The electric field is a pulse with 18 cycles (48 femtoseconds), where the first 2 cycles are ascending, the following 14 cycles have constant amplitude and finally the last 2 cycles are descending. The ionization energy for the helium is $I_p = 0.904^a : au.$ (24.602 eV), and for the neon is $I_p = 0.793 au.$ (21.57 eV). Only the outermost electron is considered for calculations. The ionization for both gases with above intensity is too small and is not included in the calculations.

Fig. 3.3 shows the relative phase between the gases. The relative phase of calculations has been adjusted to the range $[0, \pi]$ to be the same as the experimental data. For the rest of the figures the phase is in the range $[0, 2\pi]$. This last range permit us to know if the electron from the neon recombines after or before from the electrons of the helium. In contrast, in the range $[0, \pi]$ we lost this information about that recombines first.

The comparison between the calculations and the experimental data have an acceptable agreement in the harmonic range of $23^{th} - 41^{th}$ with the quantum path model (QP) that is calculated from Eq. (2.115). These calculations confirm the conclusion from Kanai in the sense there is a destructive interference at low harmonics when both gases are mixed. In contrast, from the 43^{th} harmonic

onward, the theoretical model reproduces strong oscillations for the relative phase not observed in experiments.

Figure 3.4: For the higher-order harmonics where there are not experimental data, we use a reference the Schrödinger solver (Qprop) $d(\omega)$ from Eq. (2.115) is replaced by $-d^*(w)$ to compare with the Fast Fourier transform of Qprop because it uses negative exponential complex

To study the part of the spectrum corresponds to the high-order harmonics where there are not experimental data, we use an Schrödinger solver Qprop[14] as reference with different atomic potentials, see Fig. 3.4. The relative phase for the harmonics at low-order is strongly dependent of the selected potential, only at the high-order harmonics all potentials reproduces the same relative phase. The atomic potential used in Fig. 3.4 is $V(r) = -1/r - e^{\alpha r}(Z-1)/r$ where the α is a coefficient that is adjusted to obtain the right I_p eigenvalue of the energy for each gas. Fig. 3.4 shows a good agreement between both calculations from the 53th harmonic onward.

The next step is to analyze whether there is any kind of structure behind the erratic oscillations of the relative phase, to determine its dependency on the length of pulse and its intensity.

We carry out several simplifications. First, we select the temporal section where the electric field has constant envelope, therefore, the movement of the electron is the same in every subcycle. The recombination and ionization times for any later subcycle are related with the first subcycle by the expression $\Re[\tau_j] = \Re[\tau_0] + jT/2$, where j is an index of the subcycle and τ_j may be the recombination or the ionization time. The complex part $\Im m[\tau_j]$ is the same for all subcycles; it is interpreted how the time spent by the electron to go through

the potential barrier during the ionization or the recombination. Therefore, according to Eq. (3.6) and the dipole value trajectories which have the ionization time in the first subcycle, $\Re[\tau_{i0}] \in [0, T/2]$, we have

$$\begin{aligned}
 d(\omega) = & d_s^1(\omega) \sum_{j=0}^{m-2} e^{i\alpha(\omega)j} + d_l^1(\omega) \sum_{j=0}^{m-3} e^{i\alpha(\omega)j} \\
 & + d_s^2(\omega) \sum_{j=0}^{m-4} e^{i\alpha(\omega)j} + d_l^2(\omega) \sum_{j=0}^{m-5} e^{i\alpha(\omega)j} \\
 & + d_s^3(\omega) \sum_{j=0}^{m-6} e^{i\alpha(\omega)j} + d_l^3(\omega) \sum_{j=0}^{m-7} e^{i\alpha(\omega)j}
 \end{aligned} \tag{3.9}$$

with $\alpha(\omega) = \frac{T\omega}{2} + \pi$. Each summation is the interference from the harmonics generated in the previous subcycles. The phase π indicates the dependency on the sign of the electric field that changes in every subcycle, m is the number of subcycles where the pulse laser has a constant envelope (each period T of the electric field has two subcycles). The upper index in the summation indicates that the short and long trajectories have different number of subcycles at the recombination. For a freed electron at $T/2$ when the trajectory is short $\Re[\tau_r - \tau_i] < T/2$ needs two subcycles: $T/2 + T/2$ from ionization to recombination. For long trajectories $\Re[\tau_r - \tau_i] \sim T$ the electron needs three subcycles: $T/2 + T$. See Fig. 3.6. For every visit to parent ion without recombination is added two subcycles more T .

The QP1 on top of Fig. 3.5 denotes the $\Delta\phi$ obtained using the quantum path which recombines in the first visit to parent ion, the first two terms from Eq. (3.9). At low harmonics $19^{th} \sim 25^{th}$, QP (full trajectories) and QP1 has similar relative phase, and also similar to the experimental data Fig. 3.3. The weight of the module of the dipole moment from the long trajectories is predominant over the short ones $|d_l^1(\omega)| \gg |d_s^1(\omega)|$ and its contribution to the module is higher than following visits $|d_l^1| > |d_l^2|, |d_l^3|$, see Fig. 3.5(b). As a result, the long trajectories d_l^1 from the helium and neon are the main contributors to relative phase.

In the middle of region $27^{th} \sim 47^{th}$ seems there is not any clear pattern. In this region we have strong oscillations as a result of interference between all trajectories. That is due to there is not a predominant dipole modulus from an specific trajectory, see Fig. 3.5(b).

Finally, from the 47^{th} harmonic onward there is a very good agreement between both models: QP and QP1, which confirms that the contribution comes

Figure 3.5: Fig. (a): Difference of phase from helium and neon. Low is the same data as Fig3.3. QP1: Selections of trajectories recombines in first visit to parent ion in Eq. (3.11). Fig(b): dipole modulus, in atomic units, when the electron recombines in same cycle $0 < \Re[\tau_r - \tau_i] < T$ first parent visit. $T < \Re[\tau_r - \tau_i] < 2T$ second parent visit and $2T < \Re[\tau_r - \tau_i] < 3T$ third parent visit. The short trajectories are plotted with small dotted, both trajectories converges at cut-off

mainly from $d_l^1(\omega)$ and $d_s^1(\omega)$. Near the cut-off appears strong oscillations as a consequence of the interference between the short and long trajectories, they have similar dipole modulus although different phase. This oscillations also appear in the Schrödinger solver, although there is not possible to explain why it happen. Once both trajectories converges near the cut-off, the oscillations disappear and remain a constructive interference $\Delta\phi \sim 0$.

At the 69th harmonic of the helium, both short and long trajectories converges to the same value. From this harmonic onward the points from the short trajectories are discarded because its has not physical meaning [87], as a consequence appears an small jump on the top figure. The same happen with neon at the 67th harmonic. This jump could be solved using the uniform approximation, see section 2.4.6

This results does not change appreciably when we modify the length of the pulse, given by the value m in Eq. (3.9). When m has a high value $d(\omega)$ can be approximated by

$$d(\omega) \simeq [d_s^1(\omega) + d_l^1(\omega)] \sum_{j=0}^{m-2} e^{i\alpha(\omega)j} \quad (3.10)$$

and the relative phase $\Delta\phi$ cancels the contribution to the phase from the last summation

$$\begin{aligned} \Delta\phi \simeq & \text{Arg}[d_s^1(\omega)_{He} + d_l^1(\omega)_{He}] \\ & - \text{Arg}[d_s^1(\omega)_{Ne} + d_l^1(\omega)_{Ne}] \end{aligned} \quad (3.11)$$

where there is not dependency on m , then only one short and one long trajectories are enough to calculate the phase while the ionization is negligible.

The relative phase in both extremes of the spectrum at lower-order and higher-order harmonics, practically does not change when the intensity of the laser increases. According to Eq. (2.115) the phase has several contributions:

$$\phi(\omega) = \Re[\omega\tau_r] - \Re[S(k_s, \tau_i, \tau_r)] + \chi(\tau_i, \tau_r) \quad (3.12)$$

where the first two terms comes from the real part of the exponential and $\chi(\tau_i, \tau_r)$ reflects the contribution of all terms before exponential where is included the diffusion term, the dipolar transition matrix, and other terms generated during the integration. Then, $\phi(\omega)$ depends on the ionization and recombination times τ_i, τ_r . At lower-order harmonics τ_i and τ_r have similar values

Figure 3.6: This figure shows what is the value of the harmonic produced when electron is set free at t_i and recombined at t_r . There is two couple of (t_i, t_r) , from long and short paths, produce the same frequency. The figure contains the times from two gases helium and neon for two intensities $3.0 \times 10^{14} \text{ W/cm}^2$ and $4.0 \times 10^{14} \text{ W/cm}^2$. The times corresponds the electron visit to the parent ion the first time. Data extracted from cycle 6 from the electric field.

independent of the intensity of the laser, see Fig. 3.6. For harmonics beyond the cutoff, τ_i and τ_r are the same for different intensities. Therefore, the increase of intensity stretched the spectra of the relative phases but remains fixed for both extremes.

Fig. 3.7 contains the relative phases for: the short quantum path, the long quantum path, and both quantum paths (QP1), for the first visit to parent ion. Figure shows QP1 oscillates around of the relative phase of long quantum paths. Only the long quantum paths are enough to predict the relative phase at both extremes of the spectrum. Therefore, when the intensity increases, the times for ionization and recombination at low-order harmonic and beyond the cutoff are very similar and as a consequence relative phase are similar at the extremes. The strong oscillations near the cutoff where relative phase oscillates between 0 and π opens the opportunity to explore this effect. The most obvious is to cancel an harmonic with a proper ratio between the gases. In fact, the harmonics where the relative phase is destructive permits to adjust the phase of the harmonic generated tuning the ratio between the gases. To study other physical processes like the continuum-continuum interaction because all calculations are based on continuum-bound transitions and this effect is removed for

Figure 3.7: The dash line is the $\Delta\phi = d_{l(He)}^1(\omega) - d_{l(Ne)}^1(\omega)$ for long trajectories in the first visit to the parent ion. The dot-dash line is for short trajectories $\Delta\phi = d_{s(He)}^1(\omega) - d_{s(Ne)}^1(\omega)$. The thick line oscillates around the dash line, and it is enough to know the relative phase in the extremes.

destructive interference.

An approximation very extended in our community, when there is a mixture of two gases, is $\Delta\phi = \Delta I_p \Delta(t_r - t_i)$, where the time corresponds to the long paths. Fig. 3.8 shows that this approximation is good at low order harmonics although, it fails at high order harmonics 53^{th} to 65^{th} in the sense it can not predict the oscillations created by interference between the long and short paths, although produces similar values to that of QP1 for the most of harmonics.

Finally, Fig. 3.9 shows the interference using several densities ratio *helium/neon*. Clearly appears two harmonics 55^{th} , 61^{th} where there is a destructive interference such as appears in Fig. 3.7 although for different ratio density for every harmonic. The 57^{th} harmonic for the helium has a very low value compared to other from its spectrum, therefore to determine any kind of interference, the density ratio have to be much higher than 30, the maximum appears in the figure. Rest of harmonics have a constructive interference.

3.4 Conclusions

The model based on the quantum paths allows to explain the constructive and destructive interference between the harmonics generated with two noble gases like helium-neon. It explains the destructive interference at low order

Figure 3.8: The simplification $\Delta I_p \Delta t$ is based on long paths. Both models has similar values at low harmonics, where module of the dipole moment, from the long paths, is predominant. At harmonics near cut-off from 53^{th} to 65^{th} where is predominant the contribution to the first visit to the parent ion, the approximation $\Delta I_p \Delta t$ fails for those harmonics where the interference is destructive, although produces result similar to that of QP1 for most of harmonics: 53^{th} , 57^{th} , 59^{th} , 65^{th}

Figure 3.9: Interference due to the mixture helium-neon for several densities ratio (He/Ne) densities: 0,10,20 and 30. Some harmonics has constructive interference compared with the case only neon, and others destructive.

harmonics and the constructive-destructive oscillations near the cutoff. This results can be extended in a wide range of intensities and the pulses length. The ratio between densities of each gas can be used for adjusting the intensity and the phase of harmonics. Perhaps the most interesting cases are those in which the relative phase is near π because with a proper ratio between the gases we can obtain a particular phase for the harmonic. Finally, the selection of the dipolar transition matrix for neon which takes into account the parity of electron, with mathematical expression different from those as the commonly used, was decisive in reproducing the experimental and numerical results from the Schrödinger solver.

Chapter 4

Laser with orthogonal polarization

4.1 Introduction

Nowadays, there is a growing interest in the area of the circularly polarized light of the soft-X-ray due to the possibilities that it offers to use this as probe for exploring the chirality of the light-matter interactions. The circularly polarized light is particularly useful for the X-ray magnetic circular dichroism (XMCD) spectroscopy. For the magnetic materials, the XMCD spectroscopy permits to extract a detailed information about the magnetic state by distinguishing between the spin and the orbital magnetic moments of each element as well as the magnetic structures with a nanometer resolution. [35][62][56]

The circularly polarized soft-X-ray has been restricted to a few large facilities, such as the free-electron lasers or the synchrotrons. An alternative solution is a compact source that is based on the high-order harmonics generation (HHG) derived from their unique ability to generate a bright, broadband, ultra-short, and coherent light. The HHG sources enable the coherent imaging of the nanometer structures, and the access to the fast dynamics.

Usually, the linearly polarized field is used to produce the high-order harmonics, however the circularly polarized field does not produce any harmonics at all. In contrast, the circularly polarized harmonics can be generated by an appropriate mixing of two-colors lasers [18, 96]. Such a scheme was first realized in the experiment by Eichmann et al. [34] who employed two circularly polarized electric fields with frequencies ω and 2ω with the electric field vectors rotating in the same plane.

From the theoretical point of view, the circularly polarization has been investigated by various groups, some of them, following the numerical resolution

of the time-dependent Schrödinger equation (TDSE) [93, 94, 75, 82]. A different approach was based on the quantum paths, whose role in the HHG and ATI was firstly discussed in the early theoretical studies by Lewenstein et al. [64],[65] and Becker et al. [20] and studied in detail by Salieres et al. [85]. Another important contributions using the circularly polarized field comes from the theoretical studies carried on by Milošević et al.[19, 72, 70, 86]. The quantum paths are the result of solving the dipole moment integral in the momentum space under the approximation of stationary phase. The solutions are referred as the saddle points and they are a set of complex quantities, whose the real parts is in agreement with the simple-man model and the complex part takes into account the tunneling ionization. A small number of the quantum paths is enough for describing the generation of the harmonics. In particular, two classes of the quantum paths provide the most relevant contribution: the so-called long and short paths whose return times are of the order of one optical-cycle, and half optical-cycle respectively.

This chapter is organized as follows. The theoretical section describes how to obtain the mathematical expressions of the dipole matrices under the SFA approximation for outermost electrons of the neon. Only the case in which the electric field is orthogonally polarized is considered, and as a consequence the expression of the dipole matrices depend only on the electrons with the magnetic quantum number $m = \pm 1$. From the calculation of the saddle points one obtains the contribution of the quantum paths for each electron with the quantum number $m = \pm 1$. To conclude this section, we deduce the mathematical formula of the dipole moment as summation of quantum paths.

The second section includes two comparisons. For one side, the spectra obtained using the new dipolar matrices are compared to the hydrogen-like dipolar transition matrix. And, for the other side, the comparison is with the spectrum of TDSE solver (Qprop) [14]. The Qprop code is an Schrödinger solver that couples the quantum number $m = \pm 1$ of the $2p$ orbitals with the electric field.

The third section analyses the contribution of the electrons with the quantum number $m = \pm 1$ to the high-order harmonics generation. Using an electric field with a bow tie shape, see in Fig. 4.6, the spectrum produced by an electron with $m = +1$ differs from the spectrum with $m = -1$; where both spectra have the odd and even harmonics. However, when the spectrum has the contribution from both electrons, $m = \pm 1$, the interference produced between them annihilates some harmonics.

Finally, a field with constant envelope generates harmonics linearly polarized on the X or Y axes, not both simultaneously. The circularly polarized harmonics require the presence of ascending or/and descending cycles in the envelope of the electric field. The constant envelope of the electric field is responsible for the intensity of the harmonics, however the part where the envelope changes is the responsible for the circular polarization. So, in our opinion, the shape of envelope of the electric field determines the ellipticity of the harmonics.

4.2 Theoretical Model

A key piece of the Lewenstein model Eq. (4.11) is the dipolar transtion matrix. The following paragraphs show how to obtain the dipolar transition matrix for the $2p$ electrons of the neon under the assumption that the electric field is orthogonally polarized. The dipolar transition matrix $\langle \mathbf{k} | \mathbf{r} | \Psi_0 \rangle$ is calculated from the wavefunction in the momentum space $\langle \mathbf{k} | \Psi_0 \rangle = F_{nl}(k) Y_{lm}(\theta, \phi)$; where both mathematical expressions are related by

$$\langle \mathbf{k} | \mathbf{r} | \Psi_0 \rangle = i \nabla_{\mathbf{k}} \langle \mathbf{k} | \Psi_0 \rangle \quad (2.98)$$

The calculation of the gradient in Eq. (2.98) uses the following mathematical equations that relates the Cartesian and spherical coordinates [44]:

$$\begin{aligned} \frac{\partial}{\partial k_z} [F_{nl} Y_{lm}(\theta, \phi)] = & \\ & \sqrt{\frac{(l+m+1)(l-m+1)}{(2l+3)(2l+1)}} Y_{l+1,m}(\theta, \phi) \left(\frac{dF_{nl}}{dk} - l \frac{F_{nl}}{k} \right) \\ & + \sqrt{\frac{(l+m)(l-m)}{(2l+1)(2l-1)}} Y_{l-1,m}(\theta, \phi) \left(\frac{dF_{nl}}{dk} + (l+1) \frac{F_{nl}}{k} \right) \end{aligned} \quad (4.1)$$

$$\begin{aligned} \left(\frac{\partial}{\partial k_x} + i \frac{\partial}{\partial k_y} \right) [F_{nl} Y_{lm}(\theta, \phi)] = & \\ & \sqrt{\frac{(l+m+2)(l+m+1)}{(2l+3)(2l+1)}} Y_{l+1,m+1}(\theta, \phi) \left(\frac{dF_{nl}}{dk} - l \frac{F_{nl}}{k} \right) \\ & - \sqrt{\frac{(l-m)(l-m-1)}{(2l+1)(2l-1)}} Y_{l-1,m+1}(\theta, \phi) \left(\frac{dF_{nl}}{dk} + (l+1) \frac{F_{nl}}{k} \right) \end{aligned} \quad (4.2)$$

$$\begin{aligned}
 & \left(\frac{\partial}{\partial k_x} - i \frac{\partial}{\partial k_y} \right) [F_{nl} Y_{lm}(\theta, \phi)] = \\
 & - \sqrt{\frac{(l-m+2)(l-m+1)}{(2l+3)(2l+1)}} Y_{l+1,m-1}(\theta, \phi) \left(\frac{dF_{nl}}{dk} - l \frac{F_{nl}}{k} \right) \\
 & + \sqrt{\frac{(l+m)(l+m-1)}{(2l+1)(2l-1)}} Y_{l-1,m-1}(\theta, \phi) \left(\frac{dF_{nl}}{dk} + (l+1) \frac{F_{nl}}{k} \right)
 \end{aligned} \tag{4.3}$$

Therefore, the components of the dipole moment in the X, Y, and Z axes according to Eq. (2.98) are:

$$\begin{aligned}
 \langle \mathbf{k}|z|\Psi_0\rangle &= i \frac{\partial}{\partial k_z} [F_{nl} Y_{lm}(\theta, \phi)] \\
 \langle \mathbf{k}|x|\Psi_0\rangle &= \frac{i}{2} \left[\left(\frac{\partial}{\partial k_x} + i \frac{\partial}{\partial k_y} \right) + \left(\frac{\partial}{\partial k_x} - i \frac{\partial}{\partial k_y} \right) \right] \\
 &\quad \times [F_{nl} Y_{lm}(\theta, \phi)] \\
 \langle \mathbf{k}|y|\Psi_0\rangle &= \frac{1}{2} \left[\left(\frac{\partial}{\partial k_x} + i \frac{\partial}{\partial k_y} \right) - \left(\frac{\partial}{\partial k_x} - i \frac{\partial}{\partial k_y} \right) \right] \\
 &\quad \times [F_{nl} Y_{lm}(\theta, \phi)]
 \end{aligned} \tag{4.4}$$

As result, the dipole moment for the case of a hydrogen-like orbital $\langle \mathbf{k}|\mathbf{r}|\Psi_0\rangle$ is

$$\begin{aligned}
 \langle \mathbf{k}|x|\Psi_0\rangle &= -i \frac{2^{19/4} I p^{5/4} k \sin(\theta) \cos(\phi)}{\pi(k^2 + 2Ip)^3} \\
 \langle \mathbf{k}|y|\Psi_0\rangle &= -i \frac{2^{19/4} I p^{5/4} k \sin(\theta) \sin(\phi)}{\pi(k^2 + 2Ip)^3} \\
 \langle \mathbf{k}|z|\Psi_0\rangle &= -i \frac{2^{19/4} I p^{5/4} k \cos(\theta)}{\pi(k^2 + 2Ip)^3}
 \end{aligned} \tag{4.5}$$

Assuming the electron reacts instantaneously to the electric field $\mathbf{E}(t)$; the spherical coordinates $\mathbf{k} = (k, \theta, \phi)$ have the following meaning: k is the modulus of kinetic momentum, ϕ is defined as $\arctg(E_y/E_x) = \arctg(k_y/k_x)$, and θ takes one of the two following values: $\theta = 0$ for linearly polarized field $(0, 0, E_z)$, and $\theta = \pi/2$ for orthogonally polarized field $(E_x, E_y, 0)$.

Transforming Eqs. (4.5) from the spherical to Cartesian coordinates

$$\begin{aligned}\langle \mathbf{k}|x|\Psi_0\rangle &= -i \frac{2^{19/4} I p^{5/4} k_x}{\pi(k^2 + 2Ip)^3} \\ \langle \mathbf{k}|y|\Psi_0\rangle &= -i \frac{2^{19/4} I p^{5/4} k_y}{\pi(k^2 + 2Ip)^3} \\ \langle \mathbf{k}|z|\Psi_0\rangle &= -i \frac{2^{19/4} I p^{5/4} k_z}{\pi(k^2 + 2Ip)^3}\end{aligned}\tag{4.6}$$

since in the vectorial form

$$\langle \mathbf{k}|\mathbf{r}|\Psi_0\rangle = -i \frac{2^{19/4} I p^{5/4} \mathbf{k}}{\pi(k^2 + 2Ip)^3}\tag{4.7}$$

The symmetry in all axis in Eq. (4.7) is as a consequence of the 1s orbital has spherical symmetry. Eq. (4.7) has widely used in the literature. For other wavefunctions with radial symmetry, quantum number $l = 0$, the dipolar transition matrix have the same expression for all axis. Following the same steps as before for the Gaussian wavefunctions, the dipolar transition matrix is

$$\langle k|\mathbf{r}|\Psi_0\rangle = i \left(\frac{1}{\pi\alpha} \right)^{3/4} \frac{\mathbf{k}}{\alpha} \exp\left(\frac{-\mathbf{k}^2}{2\alpha} \right), \alpha = 0.8Ip\tag{4.8}$$

The Slater type orbitals wavefunctions in the momentum space for the 2p orbitals of the neon are given by $\langle \mathbf{k}|\Psi_0\rangle = F_{21}(k) Y_{lm}(\theta, \phi)$. Therefore, the dipolar transition matrix from the gradient Eq. (4.4) for an electric field orthogonally polarized $\theta = \pi/2$, gives for the quantum number $m = +1$;

$$\begin{aligned}\langle \mathbf{k}|x|\Psi_0\rangle_{m=+1} &= \frac{2^{19/4} I p^{7/4} [3k^2 \sin(2\phi) - 3ik^2 \cos(2\phi) - 2ik^2 + 2iIp]}{\pi(k^2 + 2Ip)^4} \\ \langle \mathbf{k}|y|\Psi_0\rangle_{m=+1} &= -i \frac{2^{19/4} I p^{7/4} [3k^2 \sin(2\phi) - 3ik^2 \cos(2\phi) + 2ik^2 - 2iIp]}{\pi(k^2 + 2Ip)^4} \\ \langle \mathbf{k}|z|\Psi_0\rangle_{m=+1} &= 0\end{aligned}\tag{4.9}$$

and for the quantum number $m = -1$

$$\begin{aligned}
 & \langle \mathbf{k}|x|\Psi_0\rangle_{m=-1} \\
 &= \frac{2^{19/4}Ip^{7/4} [3k^2 \sin(2\phi) + 3ik^2 \cos(2\phi) + 2ik^2 - 2iIp]}{\pi(k^2 + 2Ip)^4} \\
 & \langle \mathbf{k}|y|\Psi_0\rangle_{m=-1} \\
 &= i \frac{2^{19/4}Ip^{7/4} [3k^2 \sin(2\phi) + 3ik^2 \cos(2\phi) - 2ik^2 + 2iIp]}{\pi(k^2 + 2Ip)^4} \\
 & \langle \mathbf{k}|z|\Psi_0\rangle_{m=-1} = 0
 \end{aligned} \tag{4.10}$$

Where both expressions are the conjugate complex of each other, at least formally.

Only the orbitals with the quantum number $m = \pm 1$ contributes to the dipolar transition matrix when the electric field is orthogonally polarized. The electron with $m = 0$ is for the case when the electric field is linearly polarized [40].

The dipole moment in the length gauge $H_{int}(t) = \mathbf{r} \cdot \mathbf{E}(t)$ is given by

$$\begin{aligned}
 \mathbf{d}(t_r) &= i \int_{t_0}^{t_r} dt_i \int d\mathbf{k} e^{-iS(\mathbf{k}, t_i, t_r)} \\
 &\quad \times (\langle \Psi_0 | \mathbf{r} | \mathbf{v}(\mathbf{k}, t_r) \rangle \langle \mathbf{v}(\mathbf{k}, t_i) | \mathbf{E}(t_i) \cdot \mathbf{r} | \Psi_0 \rangle)_{m=+1} \\
 &+ i \int_{t_0}^{t_r} dt_i \int d\mathbf{k} e^{-iS(\mathbf{k}, t_i, t_r)} \\
 &\quad \times (\langle \Psi_0 | \mathbf{r} | \mathbf{v}(\mathbf{k}, t_r) \rangle \langle \mathbf{v}(\mathbf{k}, t_i) | \mathbf{E}(t_i) \cdot \mathbf{r} | \Psi_0 \rangle)_{m=-1} \\
 &+ c.c.
 \end{aligned} \tag{4.11}$$

where t_i and t_r are the ionization and recombination times, $\mathbf{v}(\mathbf{k}, t)$ is the kinetic momentum that is related with the canonical momentum by the expression $\mathbf{v}(\mathbf{k}, t) = \mathbf{k} + \mathbf{A}(t)$ where $\mathbf{A}(t)$ denotes the vector potential, and $S(\mathbf{k}, t_i, t_r)$ is the quasi-classical action that describes the motion of an electron in an electric field.

$$S(\mathbf{k}, t_i, t_r) = \int_{t_i}^{t_r} \frac{[\mathbf{v}(\mathbf{k}, t)]^2}{2} + I_p dt \tag{2.96}$$

Eq. (4.11) is an extension of the Lewenstein equation [64] where appears decoupled the contribution of each electron with $m = \pm 1$. Formally, the equation implicitly assumes that the quantum number m at ionization time is the same at the recombination time, that is, the electron does not change the quantum

number m . The equation implicitly assumes that an electron with the quantum number m in the ground state before ionization time remains unchanged after the recombination. By other side, the inclusion of cross-terms to allow the values interchange of m implies the ionization of two electrons and this requires much higher laser intensities.

In Eq. (4.11), the scalar term $\langle \mathbf{v}(\mathbf{k}, t_i) | \mathbf{E}(t_i) \cdot \mathbf{r} | \Psi_0 \rangle$ refers to the ionization, and the vectorial term $\langle \Psi_0 | \mathbf{r} | \mathbf{v}(\mathbf{k}, t_r) \rangle$ refers to the recombination and also, is the responsible for the values in the X-axis differs from the Y-axis.

The Eq. (4.11) is integrated by the method of stationary phase. Therefore, we get the derivative of the action respect to all variables and equate it to zero as a result, we have a set of equations which has to be solved simultaneously. The numerical solution of Eqs. (2.111) - (2.113) produces a set of complex solutions $(\mathbf{k}_s, \tau_r, \tau_i)$ that identify a reduced set of trajectories as the most important due to its contribution to the generation of harmonics. Where $\mathbf{k}_s$ is the complex momentum, τ_i is the complex ionization time, and τ_r is the complex recombination time. The imaginary part of the time alludes to the time spend to go through the atomic potential barrier. The real part is usually the time.

Based on the excursion time, the solutions of the electron trajectories are classified as long and short. For the long paths, the electrons are freed near the maximum value of the electric field and its time in the continuum is around $\Re[\tau_r - \tau_i] \sim T$, where T is the period of the electric field. On the contrary, the short paths corresponds to the electrons ionized with lower intensity and its excursion time is $\Re[\tau_r - \tau_i] \sim T/2$. This electrons returns earlier than long paths to parent ion. Some times, the electron does not recombines in the first visit to the parent ion, when the electric field changes the sign and move the electron to its origin. The electron could recombines in the second or third visit. For visits after the first one, the value of dipole moment decreases as a consequence of the attenuation factor $1/(\tau_r - \tau_i)$ in the equations (4.14)-(4.15).

The set of complex values $(\mathbf{k}_s, \tau_r, \tau_i)$ are the same for both electrons $m = -1$ and $m = +1$ because the equations used to obtain them Eqs. (2.111)-(2.113) have not explicit dependency on m . Then, the trajectories of the electrons once they go through the potential barrier are the same.

The momentum of the saddle point $\mathbf{k}_s$ is a complex magnitude, and so, the phase $\phi = \arctg(k_y/k_x)$ is complex too. When ϕ is substituted in Eqs. (4.9)-(4.10) the results of the calculations are not conjugate each other.

To determine how the spectrum depends on m , we take the Fourier transform

of Eq. 4.11. Then, for $m = +1$ is given by

$$\begin{aligned} \mathbf{d}^+(\omega) = & i \int_{-\infty}^{\infty} dt_r \int_{t_0}^{t_r} dt_i \int d\mathbf{k} \langle \Psi_0 | \mathbf{r} | \mathbf{v}(\mathbf{k}, t_r) \rangle_{m=+1} \\ & \times \langle \mathbf{v}(\mathbf{k}, t_i) | \mathbf{E}(t_i) \cdot \mathbf{r} | \Psi_0 \rangle_{m=+1} e^{i[\omega t_r - S(\mathbf{k}, t_i, t_r)]} + cc \end{aligned} \quad (4.12)$$

and, for the quantum number $m = -1$

$$\begin{aligned} \mathbf{d}^-(\omega) = & i \int_{-\infty}^{\infty} dt_r \int_{t_0}^{t_r} dt_i \int d\mathbf{k} \langle \Psi_0 | \mathbf{r} | \mathbf{v}(\mathbf{k}, t_r) \rangle_{m=-1} \\ & \times \langle \mathbf{v}(\mathbf{k}, t_i) | \mathbf{E}(t_i) \cdot \mathbf{r} | \Psi_0 \rangle_{m=-1} e^{i[\omega t_r - S(\mathbf{k}, t_i, t_r)]} + cc \end{aligned} \quad (4.13)$$

Integrating Eq. (4.12)-(4.13) by means of stationary phase approximation, gives as a result a superposition of the following quantum paths

$$\begin{aligned} \mathbf{d}^-(\omega) = & \sum_q \left(\frac{2\pi}{i(\tau_{qr} - \tau_{qi})} \right)^{3/2} \sqrt{-2\pi i} \times \left(\frac{\partial^2 S}{\partial t_r^2} \right)^{-1/2}_{\tau_{qr}} \\ & \times e^{i(\omega \tau_{qr} - S(\mathbf{k}_{qs}, \tau_{qi}, \tau_{qr}))} \\ & \times \langle \Psi_0 | \mathbf{r} | \mathbf{v}(\mathbf{k}_{qs}, \tau_{qr}) \rangle_{m=-1} d_{ion}(q)_{m=-1} \end{aligned} \quad (4.14)$$

$$\begin{aligned} \mathbf{d}^+(\omega) = & \sum_q \left(\frac{2\pi}{i(\tau_{qr} - \tau_{qi})} \right)^{3/2} \sqrt{-2\pi i} \times \left(\frac{\partial^2 S}{\partial t_r^2} \right)^{-1/2}_{\tau_{qr}} \\ & \times e^{i(\omega \tau_{qr} - S(\mathbf{k}_{qs}, \tau_{qi}, \tau_{qr}))} \\ & \times \langle \Psi_0 | \mathbf{r} | \mathbf{v}(\mathbf{k}_{qs}, \tau_{qr}) \rangle_{m=+1} d_{ion}(q)_{m=+1} \end{aligned} \quad (4.15)$$

and the full Fourier transform is

$$\mathbf{d}(\omega) = \mathbf{d}^+(\omega) + \mathbf{d}^-(\omega) = (d_x(\omega), d_y(\omega), 0) \quad (4.16)$$

where the q index in the summation identifies the saddle point $(\mathbf{k}_{qs}, \tau_{qr}, \tau_{qi})$, the term $d_{ion}(q)$ is defined as

$$d_{ion}(q) = (-2)^{-\nu} M(\mathbf{v}(\mathbf{k}_{qs}, \tau_{qi})) J(\nu - 1, S) \left(\frac{\partial^2 S}{\partial t_i^2} \right)^{1-\nu}_{\tau_{qi}} \quad (2.124)$$

According to the definition of the wavefunction in the momentum space $\langle \mathbf{k} | \Psi_0 \rangle = F_{nl}(k) Y_{lm}(\theta, \phi)$, and its simplification when the atom is under the interaction of an electric field orthogonally polarized, is

$$\begin{aligned} \langle \mathbf{k} | \Psi_0 \rangle_{m=-1} &= \frac{2^{19/4} I_p^{7/4} k (i \sin(\phi) - \cos(\phi))}{\pi (k^2 + 2I_p)^3} \\ \langle \mathbf{k} | \Psi_0 \rangle_{m=+1} &= \frac{2^{19/4} I_p^{7/4} k (i \sin(\phi) + \cos(\phi))}{\pi (k^2 + 2I_p)^3} \end{aligned} \quad (4.17)$$

where i is the complex unit, and $(k\cos(\phi), k\sin(\phi), 0)$ is the electron velocity $(v_x(t_i), v_y(t_i), 0)$ at the ionization time.

The function $J(\nu, S)$ includes the result of the integration by parts to remove the dependence with the electric field and the solution to the singularity in Eqs. (4.9), (4.10) when $k^2 = -2I_p$, then

$$J(\nu, S) = i^\nu \sqrt{\pi} \frac{\Gamma(\nu/2)}{\Gamma(\nu)} \left(2i \left(\frac{\partial^2 S}{\partial t_i^2} \right)_{\tau_{qi}} \right)^{(\nu-1)/2} \quad (2.125)$$

Finally, $M(\mathbf{v})$ and ν comes from the splitting $\langle \mathbf{v} | \Psi_0 \rangle$ in Eq.(4.17) in two parts: the coefficient $1/(v^2 + 2I_p)^\nu$ and the rest, then $M(\mathbf{v})$ and ν for the neon are

$$\begin{aligned} M(\mathbf{v})_{m=-1} &= \frac{2^{19/4} I_p^{7/4} k(i\sin(\phi) - \cos(\phi))}{\sqrt{\pi}} \\ M(\mathbf{v})_{m=+1} &= \frac{2^{19/4} I_p^{7/4} k(i\sin(\phi) + \cos(\phi))}{\sqrt{\pi}}, \quad \nu = 3 \end{aligned} \quad (4.18)$$

4.3 The Stokes parameters

A laser with two orthogonal components $(\omega_0, 2\omega_0)$ produces an spectrum where the harmonics are circularly polarized. The X and Y axes of the harmonics draw an ellipse shape. To characterize the ellipse we calculate the minor and major axes, the rotation angle of the ellipse, and the helicity from the modulus and argument of the dipole moment spectrum. see Fig. 4.1(a). The Stokes parameters are a set of values that describe the polarization state of electromagnetic radiation. The relationship of the Stokes parameters S_0, S_1, S_2, S_3 with the modulus and argument of the dipole moment is given by

$$\begin{aligned} S_0 &= |d_x(\omega)|^2 + |d_y(\omega)|^2 \\ S_1 &= |d_x(\omega)|^2 - |d_y(\omega)|^2 \\ S_2 &= 2|d_x(\omega)||d_y(\omega)|\cos(\alpha) \\ S_3 &= 2|d_x(\omega)||d_y(\omega)|\sin(\alpha) \end{aligned} \quad (4.19)$$

where $d_x(\omega)$ and $d_y(\omega)$ are the Fourier transform of the dipole moment, which in the modulus and argument format are $d_x(\omega) = |d_x(\omega)|e^{i\phi_x}$, $d_y(\omega) = |d_y(\omega)|e^{i\phi_y}$, and $\alpha = \text{Arg}[d_y(\omega)] - \text{Arg}[d_x(\omega)] = \phi_y - \phi_x$ is the phase of dipole moment.

The ellipse parameters are related with the Stokes parameters by the equations

$$\begin{aligned}
 \Psi &= \frac{1}{2} \text{atan}\left(\frac{S_2}{S_1}\right) \\
 \epsilon &= \frac{1}{2} \text{atan}\left(\frac{S_3}{\sqrt{S_1^2 + S_2^2}}\right) \\
 \mathcal{P} &= \frac{\sqrt{S_1^2 + S_2^2 + S_3^2}}{S_0} \\
 L &= |S_1 + iS_2|, V = |S_3|, Ip = \sqrt{L^2 + V^2} \\
 a &= \sqrt{1/2(Ip + L)} \\
 b &= \sqrt{1/2(Ip - L)} \\
 h &= \text{sign}(S_3)
 \end{aligned} \tag{4.20}$$

where Ψ the angle of the rotation of the ellipse, ϵ is the ellipticity, $\mathcal{P}$ is the degree of polarization, $|V|$ is the intensity of the circular polarization, $|L|$ is the intensity of the linear polarization, Ip is the polarization intensity, a and b are the major and minor axes, and h is the helicity. The helicity is the projection of the angular momentum on the propagation direction and has two values -1 (LCP: Left circularly polarized) and 1 (RCP right circularly polarized).

One of the most important applications of the circular X-ray laser is the study of the circular dichroism. A medium with circular dichroism offers different absorption according to be the polarization of the electric field, RCP or LCP, with the frequency. Usually, the harmonics generated with a laser composed by two orthogonal fields $(\omega_0, 2\omega_0)$ have an elliptical shape instead a circular shape. An electric field elliptically polarized can be decomposed in two fields circularly polarized that rotates in counterclockwise and clockwise. So, when the helicity of elliptically polarized field is LCP,

$$a = E_L + E_R, b = E_L - E_R \tag{4.21}$$

where a and b are the axes of the ellipse, and E_L, E_R are the modulus of the electric field which rotates counterclockwise, and clockwise. When the helicity of the ellipse is RCP

$$a = E_L + E_R, b = E_R - E_L \tag{4.22}$$

The helicity of the ellipse is the same as the field E_L or E_R with higher modulus.

Figure 4.1: Fig.(a): The parameters of an ellipse: the minor and major axes, and the rotation angle are obtained from the harmonics in Eq. (4.20). Fig. (b):The harmonics with an elliptical shape can be decomposed in two circularly polarized fields which rotates with opposite directions. $E_{L(left)}$, $E_{R(right)}$

4.4 Comparison with TDSE

This section compares the results obtained from the TDSE (Qprop)[14] with those from the Lewenstein model by using the approximation of the stationary phase. One use two different dipole moment expressions for the neon based on the Lewenstein model. The first, is given by the expression of the hydrogen-like atom Eq. (4.7) and, the second uses the new Eqs. Eq. (4.10) , (4.9) that depends on the quantum number m . From now onward, all calculations that use the dipole moment given by Eq. (4.7) is referred as H-like and for those which use Eq. (4.10) -(4.9) as Ne-like. The comparison with other models, like the Gaussian (4.8) produced worst, and then, are discarded in this analyses.

The potential atomic used in Qprop is given by $V(r) = -1/r - e^{\alpha r}(Z-1)/r$, here the α is a coefficient should be adjusted to obtain the right ionization energy I_p . The computer simulation uses an electron from the level $2p$ with the quantum number $m = +1$ and $m = -1$.

The electric field has two orthogonal components: E_x and E_y . The E_x component is defined as $I = 4.0 \times 10^{14} \text{W/cm}^2$, $\lambda = 800 \text{nm}$, and a trapezoidal envelope with 2 ascending cycles, 14 cycles with constant field, and 2 descending cycles. And for E_y , $I = 1.0 \times 10^{14} \text{W/cm}^2$, $\lambda = 400 \text{nm}$, and a trapezoidal envelope with 4 ascending cycles, 28 constant cycles, and 4 descending cycles.

Due to the low intensity of the laser, the ionization is insignificant and was

not included in the calculations based on the Lewenstein model. The calculations with the Qprop, the ionization is implicitly integrated in the code, and then, is not possible to disconnect. In addition, when is used the Lewenstein model, the recombination times of the electron greater than three optical-cycles respect to the ionization time have not appreciable effects on the final results.

Figure 4.2: Spectrum of the dipole modulus $|d_x(\omega)|$ based on dipolar transition matrix Eq. (4.9)-(4.10). The black line connects the odd harmonics and the blue line connects the even harmonics. The dipole modulus of the even harmonics play a crucial role for the ellipticity.

All circularly polarized harmonics are formed by two components of the dipole moment: $d_x(\omega)$ and $d_y(\omega)$. For clarity, each figure of the spectrum of the $d_x(\omega)$, $d_y(\omega)$ have two lines that joins the odd and even harmonics separately, see Fig. 4.2. Usually, the study of the high-order harmonics under a field linearly polarized was always focused on the higher values of spectrum, given by the odd harmonics, and so, the study of the even harmonics was generally ignored because they have a insignificant value compared to the odd harmonics. In contrast, when the electric field is orthogonally polarized, the role of the even harmonics are decisive to improve the ellipticity of the harmonics. When both dipole moment components $d_x(\omega)$ and $d_y(\omega)$ have similar values the ellipticity usually could have a high value.

Figure 4.3: Dipole module in atomic units. To compare between them, H-like has been multiplied by $1/2$, and Ne-like by $1/25$. Both figures shows two lines per model. Figure (a) the top line links the odd harmonics of the spectrum and the bottom line links the even harmonics. A high degree of the ellipticity could be achieved when the line of the even harmonics from $|d_x(\omega)|$ has similar values to the line that connects the even harmonics from $|d_y(\omega)|$. The same reasoning is applied to the odd harmonics.

Fig. 4.3 shows the comparison of the dipole modulus for three models: Qprop, H-like, and Ne-like. The results from Qprop are plotted such as were generated. The results from the H-like model has been multiplied by $1/2$, and from the Ne-like has been multiplied by $1/25$, to compare them. In other calculations, with other electric field intensities, length pulses and envelope, were used the same correction-factors for comparing the spectra. Perhaps, the highest value obtained from the H-like and Ne-like is due to that a freed electron is not affected by the atomic potential. The greater correction required for the neon perhaps is as a consequence of the wavefunction in the ground state extends on space more than the hydrogen, and then, the transition $\langle \Psi_0 | \mathbf{r} | v \rangle$ produces greater values.

For $|d_x(\omega)|$ in Fig. 4.3(a), the three models reproduce similar results, although for the odd harmonics the agreement is better. The parallel lines in

the spectra of the Ne-like and H-like for the harmonics beyond the cutoff is as a consequence of using the approximation of the stationary phase. For $|d_y(\omega)|$ in Fig. 4.3(b) all models reproduce similar values up to the 54th harmonic. Some discrepancies appear beyond this harmonic. Taking the TDSE (Qprop) as reference of the highest quality calculations, the Ne-like dipole model shows better results for the odd harmonics and H-like for the even harmonics.

Figure 4.4: Comparison of the phase of the dipole moment for the models: Qprop, H-like, and Ne-like for the odd and even harmonics. Figure (a), the prediction of the Ne-like has in a very good agreement with Qprop for the odd harmonics, for the even harmonics is slightly worse. Figure (b), The Ne-like in the Y-axis and the even harmonics, the agreement with Qprop is good enough up to 62th harmonic. The H-like model shows a worse results for both axes.

Regarding the phase in Fig. 4.4, the H-like model generates different results from the Qprop and Ne-like models. This is because the orbital parity of H-like is different from Ne-like. The H-like uses the 1s orbitals and the other two models use the 2p orbitals. The dipolar transition matrix for the 1s orbitals $\langle -k|r|\Psi_{1s} \rangle = -\langle k|r|\Psi_{1s} \rangle$ and for the 2p orbitals $\langle -k|r|\Psi_{2p} \rangle = \langle k|r|\Psi_{2p} \rangle$. The comparison between the TDSE (Qprop) and Ne-like models yields a very impressive agreement with the phase ϕ_x figure (a) and the odd harmonics. The results are not so good with the phase ϕ_y and the even harmonics, although

has a acceptable agreement in the spectrum up to the 50th where the difference seems as constant phase.

In the study of the elliptical harmonics, more important than the absolute phase is the relative phase $\Delta\phi = \phi_x - \phi_y$ of every harmonic, because this is the variable used in the Stokes equations (4.20). Fig. 4.5, shows that H-like produces a good agreement with Qprop. Fig. 4.5(a) is the $\Delta\phi$ for the odd harmonics and Fig. 4.5(b) for the even harmonics.

Figure 4.5: Comparison of $\Delta\phi = \phi_x - \phi_y$. The H-like model produces very good approximations compared with the results in Fig. 4.4. The reason is the H-like Eq. (4.7) yields an internal phase shift of $\pi/2$ [40] for all harmonics in both coordinates. This phase shifting is canceled in the subtraction of the phases $\Delta\phi = \phi_x - \phi_y$.

In summary, the Ne-like model generates similar results to the TDSE (Qprop) and H-like models. Using Qprop as a reference, Ne-like is better than H-like for absolute phase calculations although it requires a larger correction for the dipole modulus. In contrast, for studying the ellipticity both H-like and Ne-like are equivalent because some parameters like: the ellipticity, the helicity, and the rotation angle of the ellipse depends only on the relative magnitudes, see in Eq. (4.20). The Ne-like model opens the possibility to determine what is the

contribution to the spectrum from the electrons with quantum number $m = \pm 1$, such as we can see in the following section.

4.5 The role of quantum magnetic numbers

This section is devoted to analyzing how the spectrum changes as a consequence of the different contribution of the electrons with $m = \pm 1$ under the interaction of the electric field such as was defined in the previous section.

Figure 4.6: Projection of the electric field on the XY-plane. The blue arrow indicates the evolution over time. For the even subcycles the field is like left circularly polarized (LCP) and for the odd subcycles is like right circularly polarized (RCP).

Fig. 4.6 shows the projection on XY-plane of the electric field from the observer point of view who can see how the field approximates to him. Only the part of the electric field with constant envelope is drawn. The figure has a bow-tie shape, where every lobe corresponds to a field subcycle. From now onward, the name *subcycle* make reference to the half of period, $T/2$, of the lower frequency from electric field, that is, the field with wave-length $\lambda = 800 \text{ nm}$. The bow-tie shape has implicitly a rotation of the electric field to the right for the odd subcycles and to the left for the even subcycles. Then, the even and odd subcycles are the mirror image to each other. In this way, the behavior of the electron with $m = +1$ when the field rotates to the left (right) should be the same as for $m = -1$ when rotates to the right (left). Therefore, both

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electrons are subjected to similar conditions after the electric field completes a full rotation on the bow-tie figure.

Figure 4.7: **E** field with $E_x : \lambda = 800 \text{ nm}$, $I = 4.0 \times 10^{14} \text{ W/cm}^2$, $E_y : \lambda = 400 \text{ nm}$, $I = 1.0 \times 10^{14} \text{ W/cm}^2$. The small digits inside the figure indicates the number of subcycle. The figure shows the frequency emitted by the harmonics at the moment of the recombination times, black lines. Only two set of trajectories short and long are plotted. Every couple of trajectories has a shape of a bell, the left side of the bell corresponds the short trajectory, the right side describes the long trajectory. The first bell corresponds to the electrons recombine in subcycle 11^{th} , that were ionized at the previous subcycle 10^{th} . The second bell is for electrons that ionize at subcycle 11^{th} and recombine at 12^{th} . The ionization times curves are not plotted in the figure. At background of the figure two curves red and blue that belong to the component X and Y from the electric field.

To simplify the study, only the quantum paths whose difference between the recombination and ionization times are lees than a period $\Re[\tau_r - \tau_i] \leq T_0$ are considered for the calculations. The ionization is not included in the calculations because it is too low for the current intensities.

By means of the quantum paths one calculates the spectrum produced by every single optical-cycle of the electric field. In the following sections we can see how is the spectrum produced by a single electron with $m = +1$ and/or $m = -1$ when the ionization takes place in a single RCP or LCP subcycle. And later on how is the spectrum produced after a complete rotation on the bow-tie in Fig. 4.6.

4.5.1 A single subcycle

One compares the spectra produced by a single electron with $m = +1$ or $m = -1$ in two cases, when the electrons are ionized in RCP or RCP subcycles. Two subcycles from the constant envelope region of the electric field are selected; the 10th subcycle for representing all LCP subcycles and the 11th subcycle for all RCP subcycles, see Fig. 4.7

Figure 4.8: Modulus of dipole moment for electrons with $m = +1$ and $m = -1$ when the electrons are ionized in a subcycle where the electric field is RCP and LCP. For the even subcycles where the field is LCP, the greater values are produced by electrons with $m = -1$. For the odd subcycles when the electric field is RCP, the greater modulus of dipole moment is produced by electrons with $m = +1$

Fig. 4.8 shows the contribution to the spectrum of the short and long quantum paths from each electron with $m = \pm 1$. The dipole moment for an electron with the quantum number $m = +1$ are $d_x^+(\omega)$ and $d_y^+(\omega)$. The dipole moment with $m = -1$ are $d_x^-(\omega)$ and $d_y^-(\omega)$. The calculations shows that the modulus of the dipole moment of the electrons with $m = -1$ is greater than $m = +1$ for electrons ionized in a LCP subcycle, and the modulus with $m = +1$ is greater than $m = -1$ for the RCP subcycles independently of the dipole

modulus component: the X or Y axis. The electrons with $m = +1$ that are ionized in the RCP subcycles has the same dipole modulus as the electrons with $m = -1$ that are ionized in the LCP subcycles. The same happen for the dipole modulus of the electrons with $m = -1$ and RCP compared to the electrons with $m = +1$ and LCP.

The following equations describes mathematically Fig. 4.8.

$$\begin{aligned} |\mathbf{d}^+(q\omega)|(LCP) &= |\mathbf{d}^-(q\omega)|(RCP) \\ |\mathbf{d}^-(q\omega)|(LCP) &= |\mathbf{d}^+(q\omega)|(RCP) \\ |\mathbf{d}^+(q\omega)|(LCP) &< |\mathbf{d}^+(q\omega)|(RCP) \end{aligned} \quad (4.23)$$

where $\mathbf{d}^+(q\omega)$ is the spectrum of $m = +1$ calculated with Eq. (4.15), and $\mathbf{d}^-(q\omega)$ is for electrons $m = -1$ using Eq. (4.14),

Figure 4.9: Phase of dipole moment of the odd harmonics. $\mathbf{d}^+(q\omega)$ corresponds to electrons with $m = +1$ and $\mathbf{d}^-(q\omega)$ is for electrons with $m = -1$, RCP, and LCP allude to the polarization of the electric field at the moment the electron is set free. Fig. (a) corresponds to the phase in the X-axis of dipole moment. Fig. (b) is in the Y-axis, dotted red curve is shifted π from blue line curve and dotted blue is shifted π from red curve only for the harmonics points

As far as the phase is concerning, in Figs. 4.9-4.10 the phase has been splitted in two parts: one with the odd harmonics and other with the even harmonics. Fig. 4.9(a) and Fig. 4.10(b) shows a simple description with two curves for the four spectra. On the contrary, Fig. 4.9(b) and Fig. 4.10(a) there are four curves, although really there are only two curves because the other two are the same curves although shifted π radians.

Figure 4.10: Same figure as Fig. 4.9 for the even harmonics.

Dipole argument $d_x(q\omega)$ in the X-axis and the odd harmonics is

$$\begin{aligned} \text{Arg}[d_x^+(q\omega)](LCP) &= \text{Arg}[d_x^-(q\omega)](RCP) \\ \text{Arg}[d_x^-(q\omega)](LCP) &= \text{Arg}[d_x^+(q\omega)](RCP) \end{aligned} \quad (4.24)$$

and for the even harmonics

$$\begin{aligned} \text{Arg}[d_x^+(q\omega)](LCP) &= \pi - \text{Arg}[d_x^-(q\omega)](RCP) \\ \text{Arg}[d_x^-(q\omega)](LCP) &= \pi - \text{Arg}[d_x^+(q\omega)](RCP) \end{aligned} \quad (4.25)$$

The argument of $d_x(q\omega)$ in the Y-axis and the odd harmonics

$$\begin{aligned} \text{Arg}[d_y^+(q\omega)](LCP) &= \pi - \text{Arg}[d_y^-(q\omega)](RCP) \\ \text{Arg}[d_y^-(q\omega)](LCP) &= \pi - \text{Arg}[d_y^+(q\omega)](RCP) \end{aligned} \quad (4.26)$$

and for the even harmonics

$$\begin{aligned} \text{Arg}[d_y^+(q\omega)](LCP) &= \text{Arg}[d_y^-(q\omega)](RCP) \\ \text{Arg}[d_y^-(q\omega)](LCP) &= \text{Arg}[d_y^+(q\omega)](RCP) \end{aligned} \quad (4.27)$$

As conclusion, for the two electrons are freed in the LCP subcycle, the electron with $m = -1$ has greater contribution to the spectrum than the electron with $m = +1$. When the electrons are freed in the RCP subcycle the predominant contribution comes from the case with $m = +1$. The freed electron with $m = -1$ in the LCP subcycle has the same modulus in the spectrum than the freed electron $m = +1$ in the RCP subcycle.

4.5.2 Two consecutive subcycles

To complete the study, one increases the number of subcycles where the electron can be ionized from the single subcycle up to two consecutive subcycles, with the end to complete a full rotation of the bow-tie. In this way, both electrons $m = \pm 1$ are under the same interaction with the electric field and should produce the same spectrum.

In Fig. 4.11 shows that the modulus of the dipole moment of the electrons with $m = +1$ are different from the electrons with $m = -1$. This is unexpected result, although paying attention to the spectra are not totally different. The modulus of the dipole moment $|\mathbf{d}^+(\omega)|$ is equal to $|\mathbf{d}^-(\omega)|$ for those frequencies that are harmonics of ω , see Fig. 4.11.

$$|\mathbf{d}^+(n\omega)| = |\mathbf{d}^-(n\omega)| \quad n : 1, 2, 3, .. \quad (4.28)$$

The black curve in Fig. 4.11(a) represents the contribution from both electrons and shows a destructive interference for the even harmonics and constructive for the odd harmonics. This results can be explained from Eq. (4.24) where the odd harmonics are the result of an constructive interference:

$$\begin{aligned} d_x(q\omega) &= d_x^+(q\omega)(LCP) + d_x^-(q\omega)(RCP) \\ &+ d_x^-(q\omega)(LCP) + d_x^+(q\omega)(RCP) \\ &= 2d_x^+(q\omega)(LCP) + 2d_x^-(q\omega)(LCP) \end{aligned} \quad (4.29)$$

and (4.25) where the even harmonics are produced by a destructive interference.

$$\begin{aligned} d_x(q\omega) &= d_x^+(q\omega)(LCP) + d_x^-(q\omega)(RCP) \\ &+ d_x^-(q\omega)(LCP) + d_x^+(q\omega)(RCP) = 0 \end{aligned} \quad (4.30)$$

Following the similar steps as before, Fig. 4.11(b) shows a destructive interference for the odd harmonics, and constructive for the even harmonics, see Eq. (4.27)-(4.26).

Figure 4.11: Spectrum of the dipole modulus obtained using two consecutive subcycles in the calculations. There are three different calculations: $\mathbf{d}^+(\omega)$ is the spectrum of electrons with $m = +1$, $\mathbf{d}^-(\omega)$ with electrons with $m = -1$, and $\mathbf{d}(\omega)$ with both $m \pm 1$. The spectrum $\mathbf{d}^+(\omega)$ is different from $\mathbf{d}^-(\omega)$. Both contains the even and odd harmonics. On the contrary, calculations with contributions $m \pm 1$ only contains the odd harmonics in the X-axis, and only the even harmonics in the Y-axis.

The modulus of dipole moment increases proportionally to the number of cycles for the harmonics with a constructive interference. For the harmonics with destructive interference, the increase of the number of cycles does not modify appreciable their small values, see in Fig. 4.12(c). Therefore, from Fig. 4.11, 4.12(c), a field with two collinear components ω , 2ω with constant envelope can not create any harmonics circularly polarized. The harmonics generated are linearly polarized in the X and Y axis due to the annihilation of the even harmonics in the X-axis and the odd harmonics in the Y-axis.

Therefore, to obtain the harmonics circularly polarized the envelope of the electric field can not be constant.

4.5.3 Comparison H-like with Ne-like

The calculations in this section assume that the electric field has a constant envelope and the ionization is insignificant.

Although the spectra of the H-like and Ne-like are similar, see Fig. 4.12(a) and Fig. 4.12(c), the way of the harmonics are generated is different in each model: H-like and Ne-like. For the H-like model, a single electron contributes to obtain the spectrum in Fig. 4.12(a). For the Ne-like model, with a similar spectrum as the H-like model, requires the participation from both electrons $m = +1$ and $m = -1$, because a single electron produces a spectrum as Fig. 4.12(b) which is very different from the other two spectra.

The comparison of the Fig. 4.12(b) with the other two Fig. 4.12(a) and Fig. 4.12(c) shows two great differences: first, the spectrum in Fig. 4.12(b) allows produce harmonics circularly polarized; in contrast, the harmonics in the other two figures are linearly polarized $|d_x(\omega)| \gg |d_y(\omega)|$. Second, the spectrum of a single electron with the quantum number $m = +1$ contains the odd and even harmonics in the X and Y axes.

The spectrum of the H-like model has been obtained using the following expression [40].

$$\begin{aligned} \mathbf{d}(\omega) = & \sum_q \left(\frac{2\pi}{i(\tau_{qr} - \tau_{qi})} \right)^{3/2} \sqrt{-2\pi i} \times \left(\frac{\partial^2 S}{\partial t_r^2} \right)^{-1/2}_{\tau_{qr}} \\ & \times e^{i(\omega\tau_{qr} - S(\mathbf{k}_{qs}, \tau_{qi}, \tau_{qr}))} \\ & \times \langle \Psi_0 | \mathbf{r} | \mathbf{v}(\mathbf{k}_{qs}, \tau_{qr}) \rangle d_{ion}(q) \end{aligned} \quad (2.123)$$

where the dipolar transition matrix $\langle \Psi_0 | \mathbf{r} | \mathbf{v}(\mathbf{k}_{qs}, \tau_{qr}) \rangle$ is given by Eq. (4.7), and $d_{ion}(q)$

Independently of type short or long trajectory, the constructive/destructive interference is produced by photons with the same frequency emitted by two consecutive subcycles, that is, separated by a half period $T/2$. We take two saddle points from two consecutive subcycles $(\mathbf{k}_{1s}, \tau_{1r}\tau_{1i})$ and $(\mathbf{k}_{2s}, \tau_{2r}\tau_{2i})$ where,

$$\begin{aligned} \Re[\tau_{2r}] &= \Re[\tau_{1r}] + T/2 \\ \Re[\tau_{2i}] &= \Re[\tau_{1i}] + T/2 \end{aligned} \quad (4.31)$$

for the recombination and for ionization times. In contrast, the complex part of the time refers to the time to go through the potential barrier and, due to the

periodicity of the field, this times is the same for both saddle points: $\Im m[\tau_{1i}] = \Im m[\tau_{2i}]$ and $\Im m[\tau_{1r}] = \Im m[\tau_{2r}]$

Figure 4.12: Spectrum of modulus of dipole moment obtained when the electrons are set free in the range of subcycles from the 5th to 30th; the region where the envelope of electric field is constant. see Fig. 4.7.

Figure (a). H-like model . Spectrum using the dipolar transition matrix Eq. (2.123). Figure (b). Ne-like model. Spectrum from a single electron $m = +1$ with the odd and even harmonics. Figure (c). Ne-like model. Spectrum with both electrons $m = +1$ and $m = -1$.

Replacing this two saddle points in Eq. (2.123), we can see how changes each term in the summation, and so, to study how is the interference between this two photons. The action $S(k, \tau_i, \tau_r)$ takes the same value for both saddle

points. The dipolar transition matrix $\langle \Psi_0 | \mathbf{r} | \mathbf{v}(\mathbf{k}_{qs}, \tau_{qr}) \rangle$ preserve the modulus, although changes the sign in the X-axis but not in the Y-axis. The electric field $E_x(t)$ changes sign in every subcycle, as a result the velocity changes in the same way. The electric field $E_y(t)$ is the same for both saddle points because has double frequency than $E_x(t)$. The term $d_{ion}(q)$ has the same value for both saddle points. As conclusion, only the real part of the complex exponential $i(\omega \Re[\tau_{qr}])$ and the sign of the dipolar transition matrix in the X-axis differs with this the two saddle points.

Making use of the equation $\Re[\tau_{2r}] = \Re[\tau_{1r}] + T/2$ and $T = 2\pi/\omega$ the exponential, for the harmonic $n\omega$

$$\mathcal{W}e^{i(n\omega \Re[\tau_{2r}])} = \mp \mathcal{W}e^{i(n\pi + n\omega \Re[\tau_{1r}])} \quad (4.32)$$

where the minus sign behind $\mathcal{W}$ is for the X-axis, and the plus sign is for the Y-axis, $\mathcal{W}$ groups all terms from the Eq. (2.123) that have the same modulus for both saddle points, the exponential corresponds to the phases that are different for both saddle points. In this way, the previous equation describes the dipole module corresponding from the second saddle point as function of the first saddle point.

The interference of two photons generated in two consecutive subcycles is obtained from the addition of $\mathcal{W}e^{i(n\omega \Re[\tau_{1r}])} \mp \mathcal{W}e^{i(n\pi + n\omega \Re[\tau_{1r}])}$. The interference in the X-axis is constructive for n odd and destructive for n even. In the Y-axis is constructive for n even and destructive for n odd.

For the Ne-like model in Eq. (4.15), the spectrum of the electrons with $m = +1$ has both harmonics: the odd and the even, because the dipolar transition matrix for $m = +1$ for RCP subcycle has different values from the LCP subcycle, and then, the modulus of dipole moment is different for two consecutive subcycles, see Fig. 4.8, so that, there is no cancellation in two consecutive subcycles such as happen with the H-like. Following similar steps as the H-like model for Eqs. (4.16)(4.15)(4.14), and taking into account that $\phi \rightarrow \pi - \phi$ in consecutive subcycles, we reach a similar expression as (4.32). This result is independent of the type of trajectory: short or long, and the recombination in the first, second, or third visit to parent ion. In this way is demonstrated the constant envelope does not produce circularly polarized harmonics.

To conclude, the spectrum of the H-like model, see Fig. 4.12, for a particular frequency, is the result of the interference between several subcycles. However, the spectrum of Fig. 4.12(c) needs the participation of both electrons with

$m = +1$ and $m = -1$, because the cancellation of harmonics is not produced by each isolated electron.

4.6 Dependence of the intensity of the harmonics with the intensity of the laser

This section is devoted to the study about how changes the modulus and the phase of the dipole moment $|d_x(\omega)|$, $|d_y(\omega)|$, $Arg[d_x(\omega)]$, $Arg[d_y(\omega)]$ for the odd and even harmonics when changes the intensity the component of electric field $E_y(t)$ remaining fixed the other component of the field: $E_x(t)$. In order to better understand the ellipticity of harmonics obtained from Eq. (4.19) -(4.20). With this end, the intensity of the X-axis of the laser with wave-length $\lambda = 800 \text{ nm}$ is fixed to $I = 4.0 \times 10^{14} \text{ W/cm}^2$, and for the Y-axis of the laser, the intensity with wave-length $\lambda = 400 \text{ nm}$ change in a range between $I = 0.02 - 1 \times 10^{14} \text{ W/cm}^2$.

First, only the electrons that are set free in a single subcycle, the 10^{th} subcycle, and whose recombination time is no longer that a period $\Re[\tau_{qr} - \tau_{qi}] < T$ are selected. Both short and long trajectories are treated together in the calculation of the contribution to the dipole modulus.

Fig. 4.13 shows the modulus of the dipole moment using a single ionization subcycle. Fig. (a) corresponds to the $|d_x(\omega)|$ and Fig.(b) is for $|d_y(\omega)|$. Both figures have a set of black points joined by a continuous white line. At the black points, the modulus of the dipole moment has minimum like the 25^{th} harmonic in Fig.(a) where Y-axis of the intensity of the laser is $I = 38 \times 10^{12} \text{ W/cm}^2$. Both figures contain the odd and even harmonics of the $|d_x(\omega)|$ and $|d_y(\omega)|$

The white line joins all points where the dipole modulus of the long trajectories are the same than the short trajectories, and is on this line where the black points are placed. Therefore, all minimums are the result of destructive interference between short and long trajectories.

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Figure 4.13: Modulus of dipole moment when changes the intensity of $E_y(t)$. The white line is formed by the points where the modulus of short and long trajectories take the same values. This line joins the points where dipole modulus has a minimum value. Fig. (a) modulus of dipole moment $|d_x(\omega)|$, Fig. (b) for $|d_y(\omega)|$

In this way, the spectrum is divided in two regions. The region below the white line, the modulus of the dipole moment of the long quantum paths has greater values than the short quantum path. In this region, the spectrum could be reproduced with a high degree of precision using only the long trajectories, and depreciate the contribution of the short trajectories. And, the region above the white line, where the major contribution to the dipole modulus comes from the short paths. The difference between modulus of the short and long path increases when the selected point is further from the white line.

When the strength of $E_y(t)$ much smaller than $E_x(t)$, the contribution to the spectrum of the long trajectories is predominant over the short trajectories. This is consistent with the fact that at the limit when $E_y(t) \rightarrow 0$, such as happen with a linearly polarized electric field, the modulus of the dipole moment of the long trajectories is predominant over the short ones.

The strange oscillation appears around the harmonic 69^{th} with the intensity

$I = 30 \times 10^{12} \text{W/cm}^2$ is a numerical effect, as a result that the program can not distinguish numerically the short from long trajectories near the cutoff, and it oscillate between both solutions.

Fig. 4.13 describes the spectrum based on a single subcycle. When the number of subcycles increases the interference produced by the dipole moment from the different recombination times provokes a spectrum with many more oscillations. But, for the odd harmonics of $d_x(\omega)$ and the even harmonics of $d_y(\omega)$ the phase is the same for a single or multiples subcycle, and the modulus escalates with the number of subcycles included in the calculation of the dipole modulus. In this way, the knowledge of dipole modulus from a single subcycle, like the 10^{th} , can be used to extend the calculation to a wide range of subcycles: $5^{th} - 30^{th}$ only multiplying the modulus by the number of subcycles and the phase remains same as in single subcycle.

4.6.1 Dependence of the ellipticity with the envelope of the pulse.

The study of the ellipticity for a particular harmonic requires to know the dipole moment of this frequency for both components: $d_x(\omega)$ and $d_y(\omega)$ so for, the odd and even harmonics. In the previous paragraphs was analyzed the odd harmonics of $d_x(\omega)$ and the even of $d_y(\omega)$, the even $d_x(\omega)$ and odd $d_y(\omega)$ harmonics, which are the result of a destructive interference, were not treated until now. To continue with the study, the contribution to the spectrum is splitted in two parts, the first one contains those part of the spectrum where the envelope of the field is constant: $5^{th} - 30^{th}$ subcycles. The second part is the region where the field increases and decreases, the $1^{st} - 4^{th}$ and $31^{st} - 35^{th}$ subcycle. And finally, a comparison with the spectrum obtained from all subcycles.

A set of harmonics are selected for the comparison: $21^{st} - 30^{th}$ for the low frequencies, $41^{st} - 44^{th}$ in the middle range, and $55^{th} - 60^{th}$ near the cutoff. The criteria to select this harmonics is to have a set of points near and far from the harmonics corresponding to the black points in Fig. 4.13 where the spectrum has the minimums.

Fig. 4.14 shows the modulus of the dipole moment for both components

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Figure 4.14: Figs. (a), (b) corresponds to the odd harmonics. Figs. (c), (d) are for the even harmonics. The legend of the harmonics appear to the top of every column together the each line. The figure shows the modulus of the dipole moment of the electrons are set free in the times corresponding to of the range $5^{th} - 30^{th}$ subcycle.

$d_x(\omega)$ and $d_y(\omega)$. The dipole modulus has been calculated using the contribution of the electron with ionization time t_i belong to the range of subcycle $5^{th} - 30^{th}$. For this subcycles the envelope of the field is constant and all cycles are equals, in this way, the spectrum produced in the first subcycle is repeated in each subcycle. As a consequence, for some harmonics take place a constructive interference Fig. 4.14(a), Fig. 4.14(d) and destructive in other Fig. 4.14(b), Fig. 4.14(c). Comparing the $|d_x(\omega)|$ with $|d_y(\omega)|$ for the odd harmonics Fig. 4.14(a), Fig. 4.14(b), the value of the Y-component is much smaller that X-component. The same happen with the even harmonics where Y-component has much higher value than the X-component. In this way, the odd harmonics are practically linearly polarized on the X axis, and the even on the Y axis. As conclusion a pulse with a constant envelope doesn't generates harmonics circularly polarized.

The dipole modulus in Fig. 4.14(a) and Fig. 4.14(d) is the same as Fig. 4.13, although multiplied by the factor 26, that is equal to the number of subcycles in the range of $5^{th} - 30^{th}$.

Figure 4.15: Dipole modulus of the electrons that are ionized in the region where the $\mathbf{E}(t)$ is not constant $1^{st} - 4^{th}$, $31^{st} - 34^{th}$ see Fig. 4.7

Fig. 4.15 shows the second part of the spectrum where the contribution to the dipole moment comes from the electrons that the ionization time t_i in the range of subcycles $1^{st} - 4^{th}$, $31^{st} - 34^{th}$ where the envelope of the field is increasing and decreasing. Comparing the figures (a) and (d) from Fig. 4.15 and Fig. 4.14(d), the modulus of the dipole moment from the increase-decrease region is around 10% of the constant region. For the figures (b) and (c) the modulus of the dipole moment from the increase-decrease region is much greater than the constant region, therefore, to have a circularly polarized harmonic is needed to have a field which envelope changes over time.

Finally, Fig. 4.16 contains the full spectrum using all subcycles, comparing this figure with the two previous: Fig. 4.14 and Fig. 4.15 some interesting conclusion are extracted. The main contribution to the modulus from Fig. 4.16(a)(d) comes mainly from the part where the field is constant. And the contribution to Fig. 4.16(b)(c) comes from the increase and decrease region in Fig. 4.15(b)(c). So, the ellipticity of the harmonics is adjusted with the envelope of the field. When the length of the pulse increases but remaining constant the length of increase and decrease envelope, the values in Fig. 4.16(a)(d) increases too and as a consequence, for any harmonic elliptically polarized the

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ratio between the minor a major axis b/a of an ellipse is reduced. Then, the harmonics move away from the circular shape.

Figure 4.16: Spectrum obtained of the electrons which are set free in all subcycles. The electric field used has the envelope : 2 ramp-up + 2 down-ramp + 14 constant cycles for $E_x(t)$ and the double amount of cycles for $E_y(t)$. The Fig(a) and Fig(d) are similar to Fig. 4.14(a) and Fig. 4.14(d), where the dipole modulus of Fig. 4.14 was produced by the electrons which ionization time is in the section of the laser pulse has a constant envelope. The Fig(b) and Fig(c) are the same as Fig. 4.15(a) and Fig. 4.15(d), where the dipole modulus of Fig. 4.15 was produced by the electrons which ionization time is in the section of the laser pulse increase and decrease its envelope.

4.6.2 Dependence of the ellipticity with the length of the pulse.

In this sections, is analyzed how the modulus of dipole moment for shorter pulses changes.

Two cases are studied, in both the increase and decrease region of the pulse remains unchanged and with the same values as the previous section. The first case the constant envelope has 8 cycles for the $E_x(t)$ and the double amount for $E_y(t)$. The second case has 2 cycles for $E_x(t)$ and 4 for $E_y(t)$.

Figure 4.17: Spectrum obtained using all subcycles where the electron is ionized. The electric field used has the envelope : 2 ramp-up + 2 down-ramp + 8 constant cycles for $E_x(t)$ and the double amount for $E_y(t)$. The Fig(a), Fig(b) corresponds to the odd harmonics Fig(c), Fig(d) are for the even harmonics.

Fig. 4.17 shows the first case, comparing the figures (b)(c) with its equivalents in Fig. 4.16, the values of the modulus of the dipole moment should be the same, because the values of the electric field between the ionization time and recombination time is the same for all cases. The difference that appears in the scale is due to the Fourier transform. The figures (b)(c) corresponds to the section where the envelope is not constant and the length of this section is the same in Fig. 4.17 and in Fig. 4.16, but the calculation of the Fourier transform uses the total length of the pulse, and then, the harmonics generated from this region have smaller values when the length of the pulse is greater. To check that, we can multiply by the ratio of the pulses length $(2 + 14 + 2)/(2 + 8 + 2)$

Comparing Fig. 4.17(a)(d) to its equivalent in Fig. 4.16, the shape of the harmonics is slightly different because the contribution from the increase and decrease in Fig. 4.15 is higher when the length of the pulse is lower.

The second case has the spectrum described in Fig. 4.18. The shape of this pulse is roughly similar to a gaussian envelope for length of pulse $16 fs$. Fig. 4.18 (b)(c) are the same as its equivalent in Fig. 4.16 and Fig. 4.17, using a proper correction factor: $18/6$ and $12/6$ respectively.

Figure 4.18: Spectrum obtained using all subcycles where the electron is ionized. The electric field used has the envelope : 2 ramp-up + 2 down-ramp + 2 constant envelope cycles for $E_x(t)$ and the double amount for $E_y(t)$. The Fig(a), Fig(b) corresponds to the odd harmonics Fig(c), Fig(d) to the even harmonics.

Fig. 4.18(a)(d) shows different shape than Fig. 4.16 and Fig. 4.17, because the part of the spectrum corresponding to the increase and decrease pulse region has similar values to the region with constant envelope electric field. The figure (a)(d) with only the contribution of the region with constant envelope can be obtained from Fig. 4.14 multiplied by a factor : $(2+14+2)/(2+2+2) \times (1/13) = 3/13$ where the first term corresponds to the correction for the Fourier transform due to different pulse length and the second $(1/13)$ comes from the different contribution to the spectrum 13 cycles vs. 1 cycle from the constant region.

4.7 Conclusions

The new expression of the dipolar transition matrix of the neon presented in this paper produces a similar spectrum as other models like the TDSE (Qprop) or the H-like Eq. (4.7) very extended in the literature. However, the Ne-like opens the opportunity to explore the role both electrons with $m = +1$ and $m = -1$ in the spectrum generation.

To compare on equal footing the behavior of the electron with $m = +1$ and

$m = -1$, an electric field with the bow-tie shape is selected, because this way, the field rotates to the left (LCP) and right (RCP). In this way, the modulus of spectrum from the electron freed in a LCP subcycle with $m = -1$ is greater than the electron with $m = +1$. Also, the electron freed in a RCP subcycle with $m = +1$ has the same modulus as the $m = -1$ freed in a LCP.

When the spectrum is calculated with an even number of subcycles, the spectrum of electrons with $m = +1$ is different from those with $m = -1$ and both spectra presented the odd and even harmonics for the X and Y axes. Although the H-like model produces a similar spectrum as the Ne-like model, the way that it is produced is different. The Ne-like requires the participation of both electrons $m = \pm 1$ to annihilate some harmonics.

Fig. 4.13 shows that there are some points where the emission of the harmonics can be annihilate with an unfortunately selection of intensities.

Finally, to obtain the circularly polarized harmonics, is needed to have an electric field whose envelope changes over time because when the envelope is constant, the harmonics are linearly polarized only on the X or Y axis.

Chapter 5

Interaction of a laser with a gas jet

5.1 Introduction.

I have left for the end the interaction of a laser with a gas since it seems the logical evolution after having seen how was the interaction with an atom. Although originally it was not so. This chapter corresponds to the first half of the time devoted to this doctorate and the two previous chapters was the second part. It is important to know this, because otherwise it is incomprehensible to understand why a TDSE code has been used instead of the Lewenstein model much faster, and maybe other doubts that could appear to the reader.

This chapter provides the theoretical background and an overview of the numerical methods used to simulate the propagation of a high-intensity laser pulse through a noble gas. In our case the gas used is neon, although it could be extended to any other noble gas.

To improve the intensity of the harmonics there are several possibilities. To increase the atomic emission by the combination of several lasers that with an adequate combination of the quantum path, allow to increase the intensity of some harmonics. Another option is to modify the distribution of the density of the gas using different types of medium: hollow guides, cells, arrays of the gas jets, etc. And finally, changing the shape of the laser beam, see Auguste et al.[6].

The last part of the chapter describes some calculations changing the relative position of the laser focus with the position of the nozzle, the nozzle size, the gas pressure, and the atomic potential to determine how it affects the intensity of the harmonics.

Finally the last sections show how changes over time some magnitudes in

the region of interaction between the gas and the laser.

Figure 5.1: Vacuum chamber where takes place the interaction between an infrared laser and a gas jet to generate the high-order harmonics.

5.2 The gas region.

The device where takes place the interaction between the laser and the noble gas is constituted by a vacuum chamber which contains a valve through it the gas is emitted at intervals of a few milliseconds. The vacuum chamber has connected to a pump that removes the gas expelled by the valve and prevent may enter air from the outside. The reason for making the vacuum is to remove any other gas that could interfere with the results of the experiment and ensure that the gas around the valve is removed, so that only the gas jet region contributes to the harmonic emission. The emission of the gas is not continuous, it is synchronized with the laser pulses. The size of the nozzle through which the gas leaves is of the order of 1 millimeter.

The laser cross-section in the area over the nozzle is a few microns. Then, the gas density is practically constant along the cross-section of the laser beam. By the contrary, the variation of the gas density in the direction of the propagation laser is given by a Lorentzian distribution:

$$\mathcal{N}(z) = \frac{\mathcal{N}_o}{1 + \frac{z^2}{\gamma^2}} \quad z \leq L, \gamma = FWHM/2 \quad (5.1)$$

where L is the size of the nozzle, $\mathcal{N}(z)$ denotes the density of the gas as atoms per volume, z is the axis along the propagation of the laser beam, see Fig. 5.3.

Figure 5.2: Scheme of a typical experiment of HHG, where z is the distance from the laser focus to the point of observation. r_p is the distance from the laser beam axis to the observation point. z_0 is the distance from the laser focus to the center of the valve that expels the gas. For the figure represented here, z_0 would be a negative value. The origin of coordinates system is located at the laser focus.

The origin of coordinates $z = 0$ coincides with the middle point of the valve. In the region outside the nozzle, $z > L$, the density will be zero. γ is the parameter that gives the shape of the Lorentzian distribution. This parameter corresponds to the half of the FWHM (Full Width Half Maximum), the value of the width between two points of the distribution whose value is half the maximum. $\mathcal{N}_0$ is maximum value of the density in number of atoms per unit volume that is reached in the central point of the nozzle. For example, for a pressure of 10Torr , the atoms density corresponds to $3.5 \times 10^{17} \text{atoms/cm}^3$.

Figure 5.3: Lorentzian distribution for the case of $L = 1.8 \text{ mm}$, $FWHM = 0.7 \text{ mm}$ and $\mathcal{N}_0 = 1$. The jet gas has a similar density distribution of the above function.

Usually, the value of the pressure in the experiments is very small, 10Torr or less. Bear in mind that an atmosphere is equivalent to 760Torr . For rarefied gas, the interaction between the laser and gas could be treated as the interaction between the electric field and a single atom. If the gas density were higher, an electron freed from its parent atom and pushed by the electric field could travel distance enough to reach a near atom and finally recombines with it. In this case, the spectrum generated by the atom will be more complicated due to the increase in the number of processes such as collisions and recombination with neighborhood atoms.

The density of the gas will affect the number of photons emitted. The greater the density the greater the number of photons emitted at the beginning of the pulse. However, other effects such as the refraction index, ionization, free electrons, etc. that change over time reduce the photon emission.

The intensity of the laser to generate high harmonics varies in the range of $9.0 \times 10^{13} - 1.7 \times 10^{15} \text{W/cm}^2$. When the intensity is in the low range, the electronic cloud from the atom oscillates with the laser and does not emit high-order harmonics. When the intensity reaches the top of the scale, the electric field is so intense that it cancels the atomic potential, and as a consequence, the outermost electron is released to the continuum and never returns to the primary ion, interrupting the harmonic emission from this electron. In spite of, the spectrum does not disappear, other electrons with higher ionization energy continues with the emission of the harmonics, although the intensity of the harmonics suffer an strong intensity reduction. For the intermediate intensities at the beginning of the laser pulse, the gas starts to emit harmonics, but as it gradually ionizes when the pulse reaches the last cycles, it stops emitting harmonics or emits them with a lower value.

In addition, ionization results in the appearance of free electrons in the gas, which could change the focus point where the field reaches its maximum value. see Fig. 5.22. These free electrons will modify the phase of the electric fields by means of the refractive index of the plasma. On the other hand, the number of free electrons varies from zero to the beginning of the pulse when the medium is not ionized, until a maximum value at the end when it would be highly ionized.

The lasers used to the generation of high-order harmonics have a duration of

a few femtoseconds in the range of $5 - 80 \text{ fs}$. The wavelength of these lasers are in the infrared region, usually in the range of $800 \text{ nm} - 1053 \text{ nm}$ that corresponds to titanium-sapphire or neodymium lasers.

The envelope of pulse used in simulations habitually has two forms: trapezoidal and Gaussian. The trapezoidal envelope is defined by two parameters, n_c indicates the number of cycles of the electric field with constant envelope, and n_r is the number of ascending cycles. The number of ascending and descending cycles will always be the same.

The trapezoidal shape of the field is given by the following expressions. For the area of constant amplitude:

$$\mathbf{E}(t) = \mathbf{E}_0 \sin(\omega t + \phi) \quad T = 2\pi/\omega, \quad (5.2)$$

For pulse elevation region

$$\mathbf{E}(t) = \frac{\mathbf{E}_0 t}{n_r T} \sin(\omega t + \phi) \quad (5.3)$$

For the descent region

$$\mathbf{E}(t) = -\frac{\mathbf{E}_0(t - 2n_r T - n_c T)}{n_r T} \sin(\omega t + \phi) \quad (5.4)$$

constant period $= n_c T$; pulse rise, fall $= n_r T$;

The Gaussian form is not possible to use for computational purposes because it has infinite length, so it is replaced by a function $\sin^2$ defined in a finite range how

$$D = \frac{\pi}{T n_p} \quad (5.5)$$

$$\mathbf{E}(t) = \frac{2\mathbf{E}_0}{\omega} [D \sin(Dt) \cos(Dt) \sin(\omega t + \phi) + \sin(Dt)^2 \cos(\omega t + \phi)]$$

where n_p is the number of cycles of pulse and $n_p T$ is the length of the pulse.

5.3 The laser focusing.

The laser sources usually does not reach intensities as high as $5.0 \times 10^{14} \text{ W cm}^{-2}$. These intensities are achieved by focusing with lenses or concave mirrors, as a result, the focusing modifies the wavefront of the electric field. This section will

be devoted to obtaining the analytic expression from a focused the electric field. The modulus from Eq. (5.23) and the argument from Eq. (5.22) of the focused electric field will be used to calculate the atomic dipole moment that together the atomic density will allow to calculate the nonlinear polarization, $\mathcal{P}_q^{NL}(\mathbf{r})$. Finally, in the following section 5.4 will be described how is the propagation for a single harmonic.

The propagation of the electric field is given by the Helmholtz equation which mathematical form is

$$\nabla^2 \mathbf{E} = \frac{1}{c} \frac{\partial^2 \mathbf{E}}{\partial t^2} \quad (5.6)$$

where, the electric field propagates along z-axis direction and it is orthogonally polarized respect to the propagation direction $\mathbf{n} = (x, y, 0)$

$$\mathbf{E}(x, y, z, t) = A(x, y, z) \mathbf{n} e^{i(kz - \omega t)} \quad (5.7)$$

where $A(x, y, z)$ is the envelope function that modulates the amplitude of the field. So replacing in Eq. (5.6) results

$$\left[\left(\frac{\partial^2 A}{\partial x^2} + \frac{\partial^2 A}{\partial y^2} + \frac{\partial^2 A}{\partial z^2} \right) + 2ki \frac{\partial A}{\partial z} - k^2 A \right] e^{i(kz - \omega t)} = \frac{1}{c^2} A(-\omega^2) e^{-i(kz - \omega t)} \quad (5.8)$$

and canceling terms on both sides of the equation, produces as a result

$$\left(\frac{\partial^2 A}{\partial x^2} + \frac{\partial^2 A}{\partial y^2} + \frac{\partial^2 A}{\partial z^2} \right) - 2ki \frac{\partial A}{\partial z} = 0 \quad (5.9)$$

how $\frac{\partial A}{\partial z}$ varies slowly over the distance of a wavelength, then, the envelope fulfills the following inequity

$$\left| \frac{\partial^2 A}{\partial z^2} \right| \ll \left| 2k \frac{\partial A}{\partial z} \right| \quad (5.10)$$

hence, replacing in Eq. (5.9) the result produces the known as the paraxial equation:

$$\frac{\partial^2 A}{\partial x^2} + \frac{\partial^2 A}{\partial y^2} - 2ki \frac{\partial A}{\partial z} = 0 \quad (5.11)$$

this equation has several solutions and whose profiles appear in the Fig. 5.5. The equation will not be valid if we use laser with very few periods, let say around 5 cycles, because the assumption where the envelope varies slowly with the distance of a wavelength is not fulfilled.

Then paraxial approximation supposes:

- The envelope of the pulse varies slowly compared with the wavelength.
- The field is perpendicular to the z-axis, although the field converges to a point as a consequence of the focusing, see Eq. (5.7)

Figure 5.4: This graph presents the relationship between several parameters of Eq. (5.13) and its geometric relation. The data reported in the experiments are normally confocal parameter b , and the frequency. Sometimes the focal distance is reported, and from this you can get the angle θ and all the other parameters.

When an electric field is focused, the field can be represented by an spherical wave that is attenuated with the distance:

$$\frac{e^{-ikr}}{r} = \frac{e^{ikz} \sqrt{1 + \frac{x^2 + y^2}{z^2}}}{z \sqrt{1 + \frac{x^2 + y^2}{z^2}}} \approx \frac{e^{-ikz} e^{-\frac{ik(x^2 + y^2)}{2z}}}{z} \quad (5.12)$$

The last term of equation (5.12) is obtained from the approximation $z^2 \gg x^2 + y^2$. The size of $x^2 + y^2$ is around some microns that correspond to the cross-section of the beam laser and the z size is around some millimeters. See Fig. 5.4

Replacing in Eqs. (5.12), (5.11) the solution is

$$\mathbf{E}(x, y, z, t) = \mathbf{E}_0 \frac{w_0}{w(z)} \exp \left[-\frac{x^2 + y^2}{w^2(z)} \right] \exp \left[-ik \frac{x^2 + y^2}{2R(z)} + i\zeta(z) \right] \quad (5.13)$$

$$\mathbf{E}_0 = E_0 \mathbf{n} e^{i(kz - \omega t)}$$

where $w(z)$ is the envelope function corresponding to the beam radius, $R(z)$ is the beam curvature, $\zeta(z)$ is known as the Gouy phase. These magnitudes are expressed as

$$w(z) = w_0 \sqrt{1 + \left(\frac{\lambda z}{\pi w_0} \right)^2} \quad (5.14)$$

$$R(z) = z \left[1 + \left(\frac{\pi w_0^2}{\lambda z} \right)^2 \right] \quad (5.15)$$

$$\zeta(z) = (1 + l + m) \arctg \left(\frac{z}{z_R} \right) \quad z_R = \frac{\pi w_0}{\lambda} \quad (5.16)$$

$$\Theta = 2\theta, \quad \theta = \frac{\lambda}{\pi w_0} z \gg z_R \quad (5.17)$$

where $l = m = 0$ for Gaussian beam TEM_{00} and w_0 denotes the value of the radius of the beam at the point of focus.

Figure 5.5: The spherical wave approach generates several solutions for the field equationm see Eq. (5.6). This figure illustrates the radial profiles of several possible solutions are shown

As consequence of the focusing, it is established a cone of light with an angle θ that never converges to a focal point. Near this point, the beam ceases to converge and the shape changes from a cone to a something similar to a hyperboloid of revolution, see Fig. 5.4.

When a plane wave is focused, a new phase appears. This phase was not present before when the field was described by a plane wave and it is known as Gouy phase, see Eq. (5.16). It varies in a range from $-\pi/2$ to $\pi/2$ when we move on the z -axis from $-\infty$ to ∞ .

Figure 5.6: Gouy phase produced in the envelope of the electric field as a consequence of the focusing. The phase change from $-\pi/2$ to $\pi/2$ when comes from $z \rightarrow -\infty$ to $z \rightarrow \infty$

The expression of $R(z)$ indicates how changes the wave-front curvature over z -axis. Hence, it is easy to determine how changes the wave-front from a spherical shape to plane wave at $z = 0$ point. Then, the confocal region is place where the wave-front curvature is lower. Eq. (5.13) is most simple solution from a set of possible analytical solutions. This is the solution implemented in the code Qhgh whose results appears at end of this chapter.

A more general solution of Eq. (5.6), using the Hermite-Gaussian modes is given by

$$\begin{aligned} \mathbf{E}_{l,m}(x, y, z, t) &= \mathbf{E}_0 \frac{w_0}{w(z)} H_l \left[\frac{\sqrt{2}x}{w(z)} \right] H_m \left[\frac{\sqrt{2}y}{w(z)} \right] \\ &\quad \times \exp \left[-\frac{x^2 + y^2}{w^2(z)} \right] \exp \left[-ik \frac{x^2 + y^2}{2R(z)} + i\zeta(z) \right] \\ \mathbf{E}_0 &= E_0 \mathbf{n} e^{i(kz - \omega t)} \end{aligned} \quad (5.18)$$

where $H_n(x)$ is the Hermite polynomial with the non-negative integer index n . It can be seen that with $l = m = 0$ we obtain the fundamental Gaussian beam described earlier (since $H_0(x) = 1$) Eq. (5.13). one of the possible solutions is known as the mode $TEM_{l,m} \rightarrow TEM_{00}$ (where TEM stands for transverse electro-magnetic), in which the intensity varies as a Gaussian function in the radial direction of the laser beam. The mode TEM_{00} is the selected for subsequent calculations.

Another way of writing the above expression, Eq. (5.13), in a simpler manner is

$$\begin{aligned} \mathbf{E}(r_{\perp}, z) &= \frac{b\mathbf{E}_0}{b + 2iz} \exp\left(-\frac{kr_{\perp}^2}{b + 2iz}\right) \\ k &= \frac{2\pi}{\lambda}, b = 2z_R = \frac{2\pi w_0^2}{\lambda} = n \frac{2\pi w_0^2}{\lambda_0} \end{aligned} \quad (5.19)$$

where $r_{\perp}^2 = x^2 + y^2$, λ_0 is the wavelength of central harmonic from the laser, and n is the refractive index.

The confocal parameter b , is the distance between two points whose sized is $\sqrt{2}$ of the beam waist w_0 . This parameter indicating the convergence of the beam. When b is very large, the beam will be more uniform with less transverse variations of intensity along propagation axis. In this way, the confocal parameter is roughly defined as the distance where is not possible that the intensity converges anymore, for the current focusing. The b parameter, can varies from a few millimeters to of the order of meters. Usually, the confocal parameter is greater than the size of the interaction region, in order to have the highest possible intensity in the most uniform way.

From Eq. (5.19), the modulus and argument of the electric field corresponding to the mode TEM_{00} : [39], [38] is

$$\phi(r_{\perp}, z) = \arctan\left(\frac{2z}{b}\right) - \frac{2kr_{\perp}^2 z}{b^2 + 4z^2} - kz \quad (5.20)$$

$$|E(r_{\perp}, z)| = \frac{b|E_0|}{(b^2 + 4z^2)^{1/2}} \exp\left(-\frac{kr_{\perp}^2 b}{b^2 + 4z^2}\right) \quad (5.21)$$

For a medium different from the vacuum, the wavenumber k takes the complex form $k = \Re[k] + i\Im[k]$, where the real part will refer to the phase propagation, whereas the imaginary part corresponds to an absorption/emission by the medium. Then, the above expression in Eq. (5.21) continues being valid changing $|E_0| \rightarrow |E_0 e^{-ikz}|$ to reflect the absorption by the medium.

When k changes over the propagation trajectory of the beam laser, the product kz is replaced by the integral $\int_{-\infty}^z k(z') dz'$ [61], [39].

In some cases instead of use $k(z)$ is used the refractive index which is related with the wave-number as $k(z) = n(z)\omega/c$. Then, Eq. (5.20) (5.21) are replaced

by

$$\phi(r_{\perp}, z) = \text{atan}\left(\frac{2z}{b}\right) - \frac{2\Re[k(r_{\perp}, z)]r_{\perp}^2 z}{b^2 + 4z^2} - \int_{-\infty}^z \Re[k(r_{\perp}, z')]dz' \quad (5.22)$$

$$|E(r_{\perp}, z)| = \frac{b|E_o|}{(b^2 + 4z^2)^{1/2}} \exp\left[\int_{-\infty}^z \Im[k(r_{\perp}, z')]dz'\right] \exp\left[-\frac{\Re[k(r_{\perp}, z)]r_{\perp}^2 b}{b^2 + 4z^2}\right] \quad (5.23)$$

Although it seems that k does not depend on $r_{\perp}$ because the gas density is practically constant in the cross section of the laser beam, the ionization and the intense intensity gradients modify the wave number.

Not all products kz from Eq. (5.20) (5.21) are replaced by an integral, only the last term of the phase which comes from $\mathbf{E}_0$ in Eq. (5.13). The other terms are the result of internal operations to obtain the modulus and argument from Eq. (5.19)

5.4 The electric field propagation

The propagation of a electric field in a medium with polarization $\mathbf{P}$ is governed by the following differential equation

$$\nabla^2 \mathbf{E}(\mathbf{r}, t) - \frac{1}{c^2} \frac{\partial^2 \mathbf{E}(\mathbf{r}, t)}{\partial t^2} = \mu_0 \frac{\partial^2 \mathbf{P}(\mathbf{r}, t)}{\partial t^2} \quad (5.24)$$

For the particular case where the electric field is linearly polarized $\mathbf{E}(t) = (0, 0, E_z(t))$, the polarization will be linearly polarized too $\mathbf{P}(t) = (0, 0, P_z(t))$.

The decomposition in harmonics of $E_z(t)$ and $P_z(t)$ is

$$E_z(\mathbf{r}, t) = \sum_q \mathcal{E}_q(\mathbf{r}) e^{-iq\omega t} + c.c., \quad P_z(\mathbf{r}, t) = \sum_q \mathcal{P}_q(\mathbf{r}) e^{-iq\omega t} + c.c. \quad (5.25)$$

where $\mathbf{r} = (x, y, z)$ and c.c. denotes complex conjugated terms.

Replacing Eq. (5.25) in Eq. (5.24) and equating each term with the same exponential complex $e^{-iq\omega t}$, every term satisfies the following equation

$$\nabla^2 \mathcal{E}_q + \left(\frac{q\omega}{c}\right)^2 \mathcal{E}_q = -\frac{1}{\epsilon_0} \left(\frac{q\omega}{c}\right)^2 \mathcal{P}_q \quad q : 1, 2, \dots \quad (5.26)$$

where $\mathcal{P}_q = \mathcal{P}_q^L + \mathcal{P}_q^{NL}$ is the polarization for the q^{th} harmonic of the external field $\mathcal{E}_q(\mathbf{r})e^{-iq\omega t}$ and $\epsilon_0\mu_0 = 1/c^2$. The polarization can be decomposed in a linear term $\mathcal{P}_q^L$, which depends linearly on the field and is parallel to it, and another nonlinear term $\mathcal{P}_q^{NL}$ which depends on the field as powers $\mathcal{E}_q^n$ with $n > 1$,

$$\mathcal{P}_q^{NL} = \epsilon_0 \mathcal{N} (\chi_q^{(2)} \mathcal{E}_q^2 + \chi_q^{(3)} \mathcal{E}_q^3 + \chi_q^{(4)} \mathcal{E}_q^4 + \dots) \quad (5.27)$$

The nonlinear term appears for lasers with high intensities and it is responsible for the generation of high-order harmonics. For low intensity lasers, the nonlinear term of polarization can be neglected and the linear term is sufficient to model the medium.

The linear term of the polarization depends linearly on the electric field, not on its powers, and oscillates with the same frequency as the electric field. So, it can establish a relation between the electric field and the linear polarization through the first order susceptibility $\chi_q^{(1)}$ as

$$\mathcal{P}_q^L = \epsilon_0 \mathcal{N} \chi_q^{(1)} \mathcal{E}_q \quad (5.28)$$

where $\mathcal{N}$ denotes the number of atoms, and $\chi_q^{(1)}$ is the atomic susceptibility. It will be obtained from the atomic code Qprop.[13].

The susceptibility that appears here is a scalar magnitude because external field and polarization are parallel each other and to the z-axis. When both magnitudes $\mathcal{E}_q, \mathcal{P}$ are complex and with different phases, a temporal delay appears in the atomic response to the external field. Then, to coupling both magnitudes the susceptibility has to be complex and its phase will be the phase difference between $\mathcal{E}_q$ and $\mathcal{P}$.

Replacing Eq. (5.28) in Eq. (5.26)

$$\nabla^2 \mathcal{E}_q + k_q^2 \mathcal{E}_q = -\frac{1}{\epsilon_0} \left(\frac{q\omega}{c} \right)^2 \mathcal{P}_q^{NL} \quad (5.29)$$

where k_q is wavenumber for the harmonic $q\omega$ which is defined as

$$k_q = \frac{q\omega}{c} (1 + \mathcal{N} \chi_q^{(1)})^{1/2} \approx n_q \frac{q\omega}{c} = \frac{q\omega}{v} \quad (5.30)$$

the linear term of the polarization has been included in the coefficient k_q , given as a result a simpler equation (5.29) in which only appears the nonlinear term. The term $n_q = (1 + \mathcal{N} \chi_q^{(1)})^{1/2}$ is the refractive index at frequency $q\omega$, and it is a complex quantity.

Figure 5.7: Differences of phase for a q^{th} harmonic produced as a consequence of the interaction of the electric field with several atoms along the propagation direction of the field.

For short pulses, the frequency of the laser is not unique (the Fourier transform of a finite signal), hence k_1 refers to the central frequency of the pulse.

In a non polarized medium, like the vacuum, the wavenumber k_q and the fundamental k_1 are related by $k_q = qk_1$. By the contrary, in a polarized medium, the previous equality is not generally true. When the medium is homogeneous, both wavenumber k_q , k_1 are constant.

When plane wave propagates along a medium and reaches an atom, this starts emitting harmonics. Selecting one of this harmonics, for example the q^{th} harmonic, and two different points z_1 , z_2 along the propagation axis, when the fields emitted by each atom arrive at z_p point they have different phase, see Fig. 5.7. Therefore, the presence of several sources of harmonics causes an interference between harmonics, which could be destructive. This property is known as phase-matching and the mathematical expression to study it is given by the equation $\Delta k_q = k_q - qk_1$. This expression is obtained calculating the difference of phase for the q^{th} harmonic between z_1 and z_2 at point z_p , with the following simplifications, $z_2 = 0$ and $z_1 = z_p$. The study of the phase matching is a key piece to improve the efficiency in the generation of harmonics in a gaseous medium. There are several studies about the phase-matching, some times changing the focusing geometry Aguste et al. [6], using hollow fibers T. Popmintchev [79] [78] the contribution of short/ long trajectories and beyond cutoff from Vozzi et al. [98] or the pioneering from Balcou [9] [9] [10]

The non-linear components of the polarization $\mathcal{P}_q^{NL}$ for the q^{th} harmonic can be obtained from the atomic dipole moment by means

$$\mathcal{P}_q^{NL}(\mathbf{r}, t) = 2\mathcal{N}(\mathbf{r}) \mathbf{d}_q(\mathbf{r}, t) \quad (5.31)$$

where $\mathcal{N}(\mathbf{r})$ is the atomic density and $\mathbf{d}_q(\mathbf{r}, t)$ is the q^{th} harmonic component of the dipole moment.

The dipole moment is an atomic magnitude obtained from the resolution of the equation of Schrödinger for an atom that interacts with an electric field. This is defined as

$$\langle \mathbf{d}(t) \rangle = -\langle \Psi(t) | \mathbf{r} | \Psi(t) \rangle \quad (2.90)$$

for any kind of polarization of the electric field.

Taking its spectral decomposition

$$\mathbf{d}(t') = \sum_{q=0}^{\infty} \mathbf{d}(q\omega) e^{-iq(\omega t + \phi)} + c.c. \quad (5.32)$$

where phase ϕ has been added to the spectral decomposition and it refers to the initial shift of the electric field of the plane-wave $E_z(\mathbf{r}, t) = E_{oz} e^{-i(\omega t - k_z z + \phi)}$. In this way, a shift phase in the electric field does not requires to recalculate the full dipole moment, this phase is added as a complex exponential $d(q\omega) \rightarrow d(q\omega) \exp(-iq\phi)$. Although here ϕ is the initial phase of the field, the phase ϕ can include more terms like the Gouy phase, the phase related with the propagation of the field, etc. Eq. (5.22). A detailed study from Simin and Winful [99] provides a simple intuitive explanation of the physical origin of the Gouy phase shift.

The product $q\phi$ comes from considering the q -order power of the field E^q and $t' = t + \phi/\omega$. The modulus in Eq. (5.23) and the argument in Eq. (5.22) of the previous section are used to calculate the dipole moment.

The subsequent developments are done with the following simplifications:

- The electric field is linearly polarized.
- $\mathcal{P}_q^{NL}(\mathbf{r}, t)$ is a local magnitude that depends only on the intensity of the laser $\mathbf{E}_1(\mathbf{r}, t)$ of the fundamental harmonic. The other harmonics of the field have much smaller intensity than the fundamental one. The harmonics generated by the gas atoms are too small in intensity to contribute significantly to the spectrum.
- Paraxial optics is applied. The field oscillates orthogonally to the z -axis. The envelope of the laser pulse varies slowly along of the wavelength.

Figure 5.8: The red curve corresponds to the variation of the phase of the dipole moment in radians unit for the 39th harmonic, the blue curve is for the modulus of dipole moment in Cm units. The cutoff region is below $2.1 \times 10^{14} W cm^{-2}$. In the cutoff region the phase varies smoothly and linearly it has a contribution of a single electron trajectory. In the plateau region ($> 2.1 \times 10^{14} W cm^{-2}$) the phase varies faster than the previous region of the cut-off. Two electron trajectories contribute to this phase. This behavior is easier to understand under the Lewenstein model. Calculations obtained with Qprop

Figure 5.9: The cloud represents the interaction region where is produced the new harmonics. The coordinates with quotes represents the observation point where is located the detectors.

The analytical solution for Eq. (5.29) using the Green functions is given by the following expression [49]

$$\mathcal{E}_q(\mathbf{r}') = \frac{1}{4\pi\epsilon_0} \left(\frac{q\omega}{c} \right)^2 \int \frac{e^{ik_q R}}{R} P_q^{NL}(\mathbf{r}) d^3\mathbf{r} \quad (5.33)$$

The complex exponent is positive when the propagation of the electric field goes from the left side to the right side, that is, in the direction where z increases.

The complex exponent takes the negative sign when the electric field propagates for z decreasing. The integral Eq. (5.33) extends to the region where the atoms emit the harmonic $q\omega$.

Due to the region of interaction is of the order of some millimeters and the detectors can be located several meters away, it can use the far field approximation $R \sim r' |z' - z| \gg |x' - x|, |y' - y|$,

$$\mathbf{R} = |\mathbf{r}' - \mathbf{r}| = (z' - z) \sqrt{1 + \frac{(x' - x)^2}{(z' - z)^2} + \frac{(y' - y)^2}{(z' - z)^2}} \quad (5.34)$$

the expansion of the square root as a Taylor series:

$$a \rightarrow 0, \quad \sqrt{1 + a} = 1 + \frac{a}{2} - \frac{a^2}{8} + \dots (Taylor) \quad (5.35)$$

produces as a result

$$\mathbf{R} = |\mathbf{r}' - \mathbf{r}| \sim z' - z + \frac{(x' - x)^2 + (y' - y)^2}{2(z' - z)} \quad (5.36)$$

Therefore, replacing in Eq. (5.33)

$$E_q(\mathbf{r}') = \frac{1}{4\pi\epsilon_0} \left(\frac{q\omega}{c} \right)^2 \int \frac{P_q^{NL}(\mathbf{r}) e^{-i\Delta k_q z}}{z' - z} \times \exp \left\{ \frac{ik_q [(x' - x)^2 + (y' - y)^2]}{2(z' - z)} \right\} d^3\mathbf{r} \quad (5.37)$$

above Eq. (5.37) includes the following magnitude replace

$$P_q^{NL}(\mathbf{r}) = \mathcal{P}_q^{NL}(\mathbf{r}) e^{iqk_1 z} \quad (5.38)$$

where is decoupled for one side the part that corresponds to the polarization generated in a point $\mathcal{P}_q^{NL}(\mathbf{r})$, from the initial phase $e^{iqk_1 z}$ that makes reference to the phase-shift due to the propagation of the electric field of the laser in a different medium from the vacuum $k_1 z$. The factor q from the exponential comes from the harmonic order produced by the dipole $q\omega$. How the reaction of the atom to the laser is instantaneous no more phases are included.

$$E_q(\mathbf{r}') = \mathcal{E}_q(\mathbf{r}') e^{-ik_q z'} \quad (5.39)$$

this equation includes the phase due to the propagation of the harmonic $q\omega$ from the source to the observation point. [39] [66]

Eq. (5.37) $\Delta k_q = k_q - qk_1$ reflects the phase matching for q^{th} harmonic. A single laser beam can be easily described by a system with cylindrical symmetry. To take advantage of that, is changed from Cartesian coordinates to cylindrical coordinates $\mathbf{r} = (x, y, z)$, $\mathbf{r} = (r_\perp, \theta, z)$

$$\begin{aligned} \mathbf{E}_q(\mathbf{r}') &= \frac{1}{4\pi\epsilon_0} \left(\frac{q\omega}{c} \right)^2 \int \frac{\mathcal{P}_q^{NL}(\mathbf{r}) e^{-i\Delta k_q z}}{z' - z} \\ &\times \exp \left[\frac{ik_q(r_\perp^2 + r_\perp'^2 - 2r_\perp r_\perp' \cos\theta)}{2(z' - z)} \right] r_\perp dr_\perp d\theta dz \end{aligned} \quad (5.40)$$

How $\mathcal{P}_q^{NL}(\mathbf{r})$ acquires the cylindrical symmetry of the field, it is possible to reduce the number of variables of the integral from three $(r_\perp, \theta, z)$ to two $(r_\perp, z)$ by means of the the functions of Bessel J_0 that includes the result of the integration over the θ variable.

$$\begin{aligned} \mathbf{E}_q(\mathbf{r}') &= \frac{1}{4\pi\epsilon_0} \left(\frac{q\omega}{c} \right)^2 \int \frac{\mathcal{P}_q^{NL}(r_\perp, z) e^{-i\Delta k_q z}}{z' - z} \\ &\times \exp \left[\frac{ik_q(r_\perp^2 + r_\perp'^2)}{2(z' - z)} \right] J_0 \left(\frac{k_q r_\perp r_\perp'}{z' - z} \right) 2\pi r_\perp dr_\perp dz \end{aligned} \quad (5.41)$$

So far, it has been assumed that wavenumber k_q and the refractive index n_q are constant in the interaction region. Habitually, both magnitudes depends on the space and time. There are multiple factors that modify the refractive index: the gas density changes as a Lorentzian shape, a laser is focused and its intensity varies along the propagation, generation of free electrons, gas ionization, etc. As a result, the phase of the field for an instant of time, is no longer given by the expression $E_0 e^{-i(\omega t - k z)}$ it should be replaced by the more complex expression $E_0 e^{-i(\omega t - \int_{-\infty}^z k(r_\perp, z') dz')}$ in which reflects the dependency of wavenumber with the position. In Eq. (5.41) has to be replaced by

$$\Delta k_q z \rightarrow \int_{-\infty}^z \Delta k_q(r_\perp, z') dz' = \int_{-\infty}^z k_q(r_\perp, z') - qk_1(r_\perp, z') dz' \quad (5.42)$$

and then, for the polarization term

$$\begin{aligned} \mathcal{P}_q^{NL}(r_\perp, z) &= 2\mathcal{N}(z - z_0) \mathbf{d}(q\omega) e^{-iq\phi'(r_\perp, z)} \\ &= 2\mathcal{N}(z - z_0) \mathbf{d}(q\omega) \exp \left[-iq \operatorname{atan} \left(\frac{2z}{b} \right) + i \frac{2qk_1(r_\perp, z) r_\perp^2 z}{b^2 + 4z^2} \right] \end{aligned} \quad (5.43)$$

where ϕ' is the same function as ϕ from Eq. (5.22) excluding last term which is included in Eq. (5.42).

Finally using the spectrum of the dipole moment obtained from atomic calculations from Eq. (2.90)

$$\begin{aligned}
 \mathbf{E}_q(\mathbf{r}') = & \frac{1}{4\pi\epsilon_0} \left(\frac{q\omega}{c} \right)^2 \int \frac{\mathcal{N}(z - z_0) \mathbf{d}(q\omega)}{z' - z} \exp \left[-i \int_{-\infty}^z \Re[\Delta k_q(r_\perp, z'')] dz'' \right] \\
 & \times \exp \left[-iq \operatorname{atan} \left(\frac{2z}{b} \right) + i \frac{2q \Re[k_1(r_\perp, z)] r_\perp^2 z}{b^2 + 4z^2} \right] \\
 & \times \exp \left\{ \frac{i \Re[k_q(r_\perp, z)] (r_\perp^2 + r_\perp'^2)}{2(z' - z)} \right\} J_0 \left[\frac{\Re[k_q(r_\perp, z)] r_\perp r_\perp'}{z' - z} \right] 2\pi r_\perp dr_\perp dz
 \end{aligned} \tag{5.44}$$

Where $\mathcal{N}(z - z_0)$ is the atomic density of neutral atoms Eq. (5.1), z_0 indicates the distance from the center of the gas jet to the laser spot. The factor 2 comes from the Fourier transform of the $\langle d(t) \rangle$. The last expression includes the assumption that the wavenumber is a complex magnitude. When the q^{th} harmonic is in the range of X-ray, the gas behaves as transparent without interaction with the harmonic, then k_q could be approximate by qk_0 where k_0 is the wavenumber of the fundamental harmonic in the vacuum.

So far, a general expression has been developed to calculate the field in a distant region. But at no time has an explicit expression been given to calculate the refractive index, either for the fundamental or for the q^{th} harmonic. In the next section we will discuss how to calculate both refractive indexes for the fundamental and the q^{th} harmonic.

5.5 The index of refraction.

The refractive index is a magnitude that depends on the frequency and intensity of the electric field. In this section will be studied the refractive index for two harmonics: the fundamental and q^{th} harmonic. Where the last is produced by an atom as a result of a non-linear process. There is a big difference between both cases in terms of intensity and frequency. The intensity of the q^{th} harmonics could be about 10^{-10} times smaller than of the laser however, its frequency is several tens greater. The following table shows the most important process involved in the refractive index for each case

type	fundamental harmonic	q^{th} harmonic
photo-ionization	no	yes
free electrons	yes	yes
atom polarization	yes	yes

Photo-ionization

Figure 5.10: Effective photo-ionization cross-section for the neon. For wavelengths exceeding 61 nm no absorption occurs. Source: data obtained with FAC program [42]

Most of the laser sources that generate high harmonics are in the infrared region. Usually, its wavelength is in the range of 400 nm to 1000 nm . The energy of photons from this lasers is insufficient to ionize the atom. According Fig. 5.10 the 39^{th} harmonic, whose wavelength is 20.5 nm , has an appreciable photo-ionization cross-section. Therefore, the photons corresponding to this harmonics is absorbed when they travel through the gas. As a result, two effects appear, the first is a reduction of the harmonic intensity in the detector, which depend on the number of density atoms and the cross section of the harmonic. The second effect produces an increase in the ionization of the neutral atoms. Although, the effect on the ionization can be considered insignificant, since the harmonic intensity is very small.

The low order harmonics of the laser, such as 3^{rd} , 5^{th} , 7^{th} , have enough intensity to produce a significant ionization, but its effective cross-section is zero. As conclusion, the contribution of the photo-ionization to the generation of free electrons is insignificant.

The absorption of the q^{th} harmonic is included inside wavenumber by means a complex magnitude. $\Im[k_q] = \mathcal{N}_{neut}\sigma_q/2$. where σ_q is the photo-ionization cross-section of the atom for the frequency $q\omega$. $\mathcal{N}_{neut}$ is the density of neutral atoms. The absorption length $L_{abs} = 1/\mathcal{N}_{neut}\sigma_q$. (Distance at the attenuation is $e^{1/2}$). This absorption is included in our equations as an imaginary value of wavenumber, $\Im[k_q]$, then, by multiplying it by the unity complex constant i it is obtained a attenuation factor for the refractive index.

The cross section has been obtained from the calculations with the FAC code [42]. Where it uses the expression of the weighted bound-free oscillator strength in atomic units, that is related with the photo-ionization as

$$\sigma_{pi} = 2\pi\alpha \frac{df}{dE_{h\nu}} \quad (5.45)$$

where α is the fine-structure constant, f is the oscillator strength for a transition bound-free, and $E_{h\nu}$ is the energy of the photon.

Free electrons

The free electrons appears as a result of the atom ionization. The ionization is calculated from the temporal evolution of the norm of the wave-function, which decreases over time when the intensity is sufficiently high. For intensities around of $I = 9 \times 10^{14} W/cm^2$ and wavelength $\lambda = 800 nm$ the atom is completely ionized in a few cycles of the laser. Then, when the intensity of the laser is sufficiently high, the number of free charges increase over the duration of the pulse. Therefore, in the gas two populations appear: neutral and ionized atoms. The ionized atoms still emit harmonics, although their intensity is several orders of magnitude lower than intensity of the harmonics emitted by the neutrals. Only very high intensities can produce significant intensities of the ionized atoms. In conclusion, only the neutral atoms contribute effectively to the higher-order harmonics.

Figure 5.11: Ionization: Percentage of variation of the wavefunction norm over time as a consequence of the interaction with an electric field. This figure has been obtained from the second term of the right-hand of Eq. (5.49) by Qprop code. The shape of the electric field with the 14 cycles is represented in the lower part of the figure as a black line.

The permanent presence of free charges in the gas converts in to a plasma. Knowing the density of the free electrons, the calculation of the plasma frequency is direct, and from here it is easy to obtain the refractive index of the plasma.

The plasma frequency ω_p is given by [50]

$$\omega_p(r, t) = \sqrt{\frac{en_e(r, t)}{m_e \epsilon_0}} \quad (5.46)$$

where $n_e(r, t)$ is the electron density that can change from one point to another point, and depends on the time through the ionization. m_e denotes the electron mass, and e is the electron charge. For the same intensity in two different regions, the number of free electrons from one region to another is different as the gas density changes. The other factor that modifies the electronic density is a consequence of the variation of the intensity of the electric field as a result of the laser focusing.

The modification of the refractive index as function of the plasma frequency is given by the expression

$$n(\omega, r, t) = \sqrt{1 - \frac{\omega_p^2(r, t)}{\omega^2}} \approx 1 - \frac{1}{2} \frac{\omega_p^2(r, t)}{\omega^2}, \quad \Delta n(\omega, rt) = -\frac{1}{2} \frac{\omega_p^2(r, t)}{\omega^2} \quad (5.47)$$

The free electrons have the effect of decreasing the refractive index.

Generally, the lasers used to generate high-order harmonics have a pulse length of a few femtoseconds in the range of 5 – 80 fs. This time is so short

that the electrons do not have time to recombine with the ions, so that the total population of electrons increases during the experiment. Further, neither the intensity of the laser nor the total pulse time are large enough to have appreciable movement of the electrons from one region to another in the gas.

There are several expressions to calculate the ionization rates $W[E(t)]$. In the work of D. Bauer[16] some of them are compared, together with a model based on a numerical resolution of the TDSE.

$$\Gamma(t) = \exp\left(-\int_0^t W[E(t')]dt'\right) \quad (5.48)$$

where $E(t)$ is the electric field. Where $\Lambda(t) = 1 - \Gamma(t)$ is the ionization probability.

An analytical expression of the ionization rates is Eq. (2.143) from Ammosov, Delone, and Krainov [3] This expression is implemented in a later evolution of the Qhgh code which be based on the Lewenstein model.

The other option to obtain the ionization rates is from the resolution of the Schrödinger equation, and calculate how the norm of the wavefunction changes over time, see Fig. 5.11. The difference of unity with probability of finding the electron in a small volume around the atomic nucleus gives an approximation of the ionization

$$\Lambda'(t) = 1 - \int_0^r \int_0^\pi \int_0^{2\pi} \Psi^*(r, \theta, \phi) \Psi(r, \theta, \phi) r^2 dr d\theta d\phi, \quad r \approx 5 - 10 a.u. \quad (5.49)$$

This was the option selected in the Qhgh code.

Atom polarization

When there is a distribution of positive and negative charges subject to an interaction of an electric field, the external field polarizes the charges and as a result an internal field appears that opposes the external field.

Therefore, for a neutral atom, the internal field generated by the charge displacement of the nucleus of the atom with respect to the electron cloud is proportional to the external field. So, the induced dipole has the expression

$$\mathcal{P}(t) = \epsilon_0 \chi^{(1)} \mathcal{E}(t) \quad (5.50)$$

where $\mathcal{P}$ denotes the macroscopic polarization, $\chi^{(1)}$ is the first order of the susceptibility and $\mathcal{E}$ is the electric field. Eq. (5.50) shows a linear relation between

$\mathcal{E}(t)$ and $\mathcal{P}(t)$ that is generally true for an electric field with low strength. When the intensity of the laser increases up to $1.0 \times 10^{12} \text{W/cm}^2$, the above expression is no longer valid, due to appears some nonlinear effects in the polarization.

In nonlinear optics, an extension of Eq. (5.50) can be written as a series of electric field powers as

$$\begin{aligned}\mathcal{P}(t) &= \epsilon_0 [\chi^{(1)}\mathcal{E}(t) + \chi^{(2)}\mathcal{E}^2(t) + \chi^{(3)}\mathcal{E}^3(t) + \dots] \\ &= \mathcal{P}^{(1)}(t) + \mathcal{P}^{(2)}(t) + \mathcal{P}^{(3)}(t) + \dots\end{aligned}\quad (5.51)$$

Let an electric field be defined as $\mathcal{E}(t) = Ee^{-i\omega t} + c.c.$ The second-order polarization is given by [24]

$$\mathcal{P}^{(2)}(t) = 2\epsilon_0\chi^{(2)}EE^* + (\epsilon_0\chi^{(2)}E^2e^{-i2\omega t} + c.c.) \quad (5.52)$$

where the first term is a contribution of zero-frequency and does not produce an electromagnetic field. The second term has 2ω frequency, and it is responsible for the generation of a second harmonic.

The third-order polarization using the equality $\cos^3\omega t = \frac{1}{4}\cos 3\omega t + \frac{3}{4}\cos\omega t$ is given by

$$\mathcal{P}^{(3)}(t) = \frac{1}{4}\epsilon_0\chi^{(3)}E^3\cos 3\omega t + \frac{3}{4}\epsilon_0\chi^{(3)}E^3\cos\omega t \quad (5.53)$$

where the first term describes the emission of photons whose frequency corresponds to the third harmonic. The inclusion of the higher order polarization terms increases the order of the harmonics emitted.

Taking the first three terms of Eq. (5.51) and of them only those that have the same frequency as the laser

$$\mathcal{P}(\omega)^{TOT} = \epsilon_0\chi^{(1)}\mathcal{E}(\omega) + \frac{3}{4}\epsilon_0\chi^{(3)}|E(\omega)|^2\mathcal{E}(\omega) = \epsilon_0\chi_{eff}\mathcal{E}(\omega) \quad (5.54)$$

where the second sum contains the nonlinear effects associated with the Kerr effect, dependence of the polarization with the intensity of the electric field $I = 1/2\epsilon_0 c|E(\omega)|^2$. In Eq. (5.54) the polarization varies with the electric field without considering the harmonics of higher order.

From Eq. (5.30) the refractive index as function of the susceptibility is given by

$$n = \sqrt{1 + \mathcal{N}_{neut}\chi_{eff}} \quad (5.55)$$

According to Eq. (5.54) n depends on the intensity and for many materials n is defined by the expression

$$n = n_1 + n_2 \langle \mathcal{E}^2(t) \rangle = n_1 + \alpha I n_2 \quad (5.56)$$

where n_1 represents the linear refractive index, and n_2 is a new term that includes the variation of the electric field intensity and α is a constant used to adjust the formula (5.56) with (5.54)

Figure 5.12: Shelf focusing of light

One of the processes that can occur as a result of the intensity-dependent refractive index is self-focusing, which is illustrated in the Fig. 5.12. This process can occur when a beam of light having a nonuniform transverse intensity distribution propagates through a material for which n_2 is positive. Under these conditions, the material effectively acts as a positive lens, which causes the rays to curve toward each other.

Figure 5.13: Figure (a). is the norm of the wave-function at the end of the laser pulse calculated with Qprop for a pulse with 14 cycles. Figure (b). Susceptibility in SI units. Blue line numerical values obtained with Qprop for a wide range of intensities using Eq. (5.58). Red line curve obtained with Eq. (5.57)

Figure 5.13 illustrates how changes the susceptibility with the intensity of the laser. The susceptibility increase slowly up to the intensity of $3.0 \times 10^{14} \text{ W/cm}^2$. From this point, it begins to descend. Then, Eq. (5.56), which is widely used in the literature, is not valid for high intensity lasers, because it can not predicts the decline of the susceptibility for the higher values of the intensity. The data used in the Figure 5.13 has been obtained with Qprop. The decrease in susceptibility from the maximum is due to the ionization of the atom. As a result of the ionization over time, the norm of the wave function is reduced Fig. 5.13(a), and it also reduces the density of the electronic cloud in the region near the nucleus.

To describe the behavior of the susceptibility to high intensities, a polarization function with more coefficients must be used to adjust the curve obtained with Qprop in the Fig. 5.13(b). A good agreement is obtained using the fol-

lowing expression

$$\chi_{eff} = \chi^{(1)} + 3|E(\omega)|^2\chi^{(3)} + 10|E(\omega)|^4\chi^{(5)} + 35|E(\omega)|^6\chi^{(7)} + 126|E(\omega)|^8\chi^{(9)} \quad (5.57)$$

where the susceptibility depends on some coefficients $\chi^{(1)}$, $\chi^{(3)}$, $\chi^{(5)}$, $\chi^{(7)}$, $\chi^{(9)}$ and the intensity of the electric field. Eq. (5.57) only has odd order susceptibility coefficients, [24] because the terms of second-order polarization Eq. (5.52) are independent of ω harmonic, they are function of zero-frequency and 2ω . Similar reasoning is applicable to the rest of the even powers.

The values of the susceptibility coefficients for neon in S.I. units are

type	value in SI units (neon)
$\chi^{(9)}$	$-1.2611167388389 \times 10^{-118} - 3.5449801570887 \times 10^{-116}i$
$\chi^{(7)}$	$1.3514058056562407 \times 10^{-96} + 2.519756300598978 \times 10^{-94}i$
$\chi^{(5)}$	$-4.0253564348175 \times 10^{-75} - 6.212794098588394 \times 10^{-74}i$
$\chi^{(3)}$	$4.4004650327223005 \times 10^{-54} - 4.9097415420069324 \times 10^{-52}i$
$\chi^{(1)}$	$5.866398928774512 \times 10^{-33} - 5.463497111675441 \times 10^{-30}i$

Table 5.1: Coefficients to calculate the susceptibility. This coefficients has been calculated by the case of a laser with $\lambda = 800 \text{ nm}$

The total susceptibility from the Fig. 5.13(a) has been calculated with the mathematical expression [25]

$$\chi_{eff}(q\omega) = \frac{|d(\omega)|e^{-\alpha_d i}}{\epsilon_0 |\mathcal{E}(q\omega)|e^{-\alpha_E i}} \quad (5.58)$$

where the numerator is the coefficient for the q^{th} harmonic of the Fourier transform of the dipole moment $\mathcal{F}[d(t)] = d(\omega)$ obtained with Qprop, and in the denominator the same for the electric field $\mathcal{F}[E(t)] = E(\omega)$. Thus, for the fundamental harmonic the phase α_E we take it as 0.

Eq. (5.57) used to calculate from it, the refractive index/wavenumber that is needed for the propagation of the laser along the gas jet. The high order harmonics intensity is very small, then the susceptibility is a constant. In this case the susceptibility is calculated from the results generated by Qprop for an particular harmonic using Eq. (5.58).

We are not considering the atomic susceptibility of ionized atoms, because the induced dipole is much smaller than that of the neutral atom. The coefficient

n_2 must be a positive amount to behave as a coefficient of absorption in the following equation

$$\mathbf{E} = \mathbf{E}_0 e^{i(kz - \omega t)} = \mathbf{E}_0 e^{i(\omega n_1 z/c - \omega t)} e^{-\omega n_2 z/c} \quad k = \frac{\omega(n_1 + in_2)}{c} \quad (5.59)$$

Surely, the susceptibility showed in the Fig. (5.13)(b) perhaps has higher values than those appear in the graphic. I suspect that for intensities greater than $4 \times 10^{14} \text{W/cm}^2$ the calculation of susceptibility based on an atom with a single electron produces lower values than for the case with all electrons, although it was not verified.

In the following simulations, the intensity of the laser is in the range $1 - 3 \times 10^{14} \text{W/cm}^2$ to have a small ionization and a good performance in the emission of harmonics.

5.6 Results.

The numerical results from this section has been obtained with three codes:

- Qprop for the calculation of the atomic dipole moment, the ionization and the susceptibility.
- FAC to obtain the photo-ionization cross section.
- Qhhg to the propagation of the harmonics and the laser in a gas jet.

The first two codes, FAC and Qprop are freely distributed. The Qhhg is a completely new program and it was written as part of this investigation job.

All calculations was centered at the neon atom because the time required for simulations with Qprop was huge, around 12.000 hours in a computer cluster, so, it was not feasible to use other atom. The huge amount of computing hours was due to the dipole phase. This magnitude changes very fast when the intensity of the laser increases. So, to have a curve based on the dipole phase that permits to use some kind of interpolation algorithm was needed run several thousands of simulations changing only the intensity of the laser in the range of $0.001 - 7 \times 10^{14} \text{W/cm}^2$.

The points in the previous intensity range are not equally spaced, at lower intensities, the dipole moment phase changes very slowly for those harmonics are in the cut-off region. When the harmonics are in the plateau, the dipole

phase changes faster with the intensity, see Fig. 5.8. The explanation why the phase changes faster in the plateau than in the cutoff was exposed in the previous chapters. Basically it is related with the time spent by an electron from the ionization until its recombination.

The strong phase variation for a small intensity variation could be overcome using an approximation based on phase dependence with intensity $\phi = \alpha I$ [37] [7] [103]

Where α is a constant with two values one for the plateau and other for cut-off see Fig. 5.8 where the phase of the 39th harmonic can be adjusted to this two lines. The restriction with this approach was that it reduces the possibility of using linearly or orthogonally polarized laser combinations. So Qprop was selected a general option although computationally very expensive. In the late 90's Priori et al [80] were pioneering using the Lewenstein model for the laser-gas interactions. The strong reduction in the calculation time of the Lewenstein model allowed to use for the dipole calculation an atomic model instead of an approximation.

The excessive time used by Qprop for a single simulation was the reason why was developed a new atomic code based on Lewenstein model. The new code is much faster 2 seconds versus 4 hours for a simulation with a laser of 37 fs, although the new code was not integrated with Qhhg to simulate propagation yet. In any case Qprop was not abandoned it was used as benchmark in the previous chapters.

The parameters in the simulations are based on experimental measures [84].

- Atom: Ne. Density: $\mathcal{N}_0 = 5.4 \times 10^{17} \text{atom/cm}^3 \approx 17 \text{ Torr}$. This value hasn't been correctly measured in experiments, this pressure could be in a range between 10 to 170 Torr.
- Laser:
 - The wavelength of the central laser frequency: $\lambda = 800 \text{ nm}$
 - Intensity $I = 3 \times 10^{14} \text{Wcm}^{-2}$
 - Confocal parameter $b = 5 \text{ mm}$ (roughly estimated)
 - The laser form of the envelope is constituted by 14 cycles divided in 2 rise, 10 constant, 2 descent, with a pulse of length of 37 fs.
- The harmonic studied is the 39th. Threshold intensity for this harmonic $1.1 \times 10^{14} \text{Wcm}^{-2}$. The harmonic is not emitted below this intensity.

- Nozzle size 1.8 mm, Lorentzian distribution $FWHM = 0.6$ mm,
- According the construction of the program, when z_0 is positive the laser is focused before the center of gas jet.

Figure 5.14: Figure (a): Temporal evolution of the intensity for 39^{th} harmonic. Figure (b) :Cumulative results for 39^{th} Black line : experimental, Red: calculated by Salieres [84], Blue: calculated with Qhgg code. The experimental results were obtained in collaboration with T. Ditmire and M. D. Perry at the Lawrence Livermore National Laboratory and published in the same paper.

The Fig. 5.14(a) shows how the intensity changes over time for the 39^{th} harmonic. The harmonic intensity decreases when the medium is ionized. At the beginning, in the first laser cycles, the intensity of the harmonics remains constant, but later the intensity decreases progressively as a curve similar to that of the ionization, see Fig. 5.11. Only when the laser has an intensity higher than the threshold appears the harmonic emission. In the both sides of the pulse when the intensity is below the threshold, the atoms interrupt the

harmonic emission.

The Fig. 5.14(b) illustrates the cumulative temporal results from Fig. 5.14(a). It shows a comparison between the experimental results, those calculated theoretically by Salieres and the calculations with the code of Qhhg. The Fig. is a projection of a surface on one dimension, so it represents a ring in two dimensions. This is the reason why all calculations are symmetric for the divergence from the center. The most important magnitude to compare curves is divergence. All curves reach the maximum for similar values of divergence about 8 mrad. and for all the intensity is null for 14 mrad. In the middle region from -5 mrad. to 5 mrad. the calculations do not agree with experiments. Cumulative results calculated with Qhhg have small values compared to the maximum but are not zero. The Fig. 5.14(a) displays oscillations that change over time and although at each instant of time the figure evidences several zeros the accumulated signal is never zero.

The Fig. 5.15 uses the same experimental parameters as Fig. 5.14, except for the parameter z_0 that now takes the value $z_0 = 1 \text{ mm.}$ This is the relative distance between laser focus and nozzle position. For positive values of z_0 the valve is before the laser focus. See Fig. 5.2.

The Fig. 5.15(b) shows a good agreement in the points where the divergence reaches the maximum, although the agreement is not good in the middle region nor in both sides of the figure. Comparing both Figs. 5.15(a) and 5.14(a), the Fig. 5.15(a) has less field strength in the interaction region because the focus is not centered on the valve where the gas density is higher and as a consequence the ionization is lower. So, the intensity for the 39^{th} harmonic remains almost unchanged.

The Fig. 5.16 compares the results obtained with the code Qhhg are similar to results from other calculations [84], for $z_0 = 2 \text{ mm}$ and $z_0 = 3 \text{ mm.}$ Here the calculations are far from experimental results although some possible reasons for the discrepancies could be that the pressure and the confocal parameter was not fine measured in the experiment. Although a slightly difference in the confocal parameter should not be affected too much the final results.

Figure 5.15: Figure (a) Intensity time evolution for the 39th harmonic. Figure (b): Cumulative intensity and comparison with experimental and other values obtained theoretically by Salieres [84].

Relative position of the focus laser versus the nozzle valve

The following figures are the result of several simulations where the gas pressure, nozzle size, confocal parameter and z_0 are changed. The modification of the gas pressure changes the density of the atoms and consequently the total intensity of the electric field produced by the superposition of the individual fields of the atoms. Thus, a priori one might think that the intensity of a particular harmonic could scale proportionally to the density of gas, but the increase in atomic density increases the density of free electrons, as a result of ionization, and them, these electrons reduce the intensity of the laser by means of the index of refraction. Eq. (5.47).

Figure 5.16: The figures illustrate how changes the harmonic intensity with the focus position. Figure (a) $z_0 = 2 \text{ mm}$, Figure (b) $z_0 = 3 \text{ mm}$. Both theoretical calculations Qhgh and Salieres have similar results although far from the experimental results.

The Fig. 5.17 displays that all negative values of z_0 have smaller harmonic intensities than all positive values. The profile of the red curves indicates that there is no region at the divergence angle where the harmonic intensity has clearly greater values than the others.

By the contrary, the blue lines has the maximum at 0 mrad. for $z_0 = 2 \text{ mm}, 3 \text{ mm}$ and at 6 mrad. for $z_0 = 1 \text{ mm}$. The transition from the two peaks at $z_0 = 0 \text{ mm}$ to a single peak at $z_0 = 3 \text{ mm}$ occurs progressively as z_0 increases and is a consequence of the phase-matching. The maximum values of the harmonic intensity is reached at the range $z_0 = 0 \text{ mm}$ to $z_0 = 2 \text{ mm}$ and subsequently decrease strongly for greater values than $z_0 > 2 \text{ mm}$

Dependency of the intensity of 39^{th} with the nozzle size

The Fig. 5.18 indicates how changes the 39^{th} harmonic intensity with the nozzle size. The Fig. 5.18(a) presents that the maximum are reached at the same divergence point independently of the nozzle size. The highest value is not reached with the higher length nozzle. The same happen for the Fig. 5.18(b), however, the Fig. 5.18(c) shows that the intensity is higher for the greater nozzle size. So, the best case the Fig. 5.18(a) and the Fig. 5.18(b) is not the best for the Fig. 5.18(c)

As conclusion, there is no simple rule to explain when the harmonic intensity is greater. There are several interrelated variables such as ionization, intensity of the laser and phase matching that make it difficult to predict when the

Figure 5.17: Comparison of the intensity for 39th harmonic changing the position of the laser focus, after and before the nozzle position. Blue lines is for positives values of z_0 and red lines negative values z_0

harmonic intensity have the greater value.

Dependency with the atomic potential

This paragraph compares the intensity of the 39th harmonic obtained by using two different atomic potentials with the code Qprop.

Originally, all calculations were made with the parametric potential Eq. (5.60). This potential generates for the ionization energy of neon $I_p = -0.7au$ slightly lower than that used in the literature.

Another atomic potential used within Qprop is given by Eq. (5.61). This latter potential produces a value for the ionization energy that is consistent with that published in the literature. Once both potentials were included, it might be interesting to compare simulations between them.

The label HF in Fig. 5.19 is a parametric potential obtained by the Hartree-Fock method, the general form of this expression is the following [83]

$$V(r) = -\frac{1}{r} \left((\mathcal{N}_e - \frac{1}{2})(a_0 e^{-a_1 r} + (1 - a_0)a_2 r) + Z - \mathcal{N}_e + 1 \right) \quad (5.60)$$

where Z denotes the nuclear charge, $\mathcal{N}_e$ is the number of electrons, and a_* is a set of parameters given in tables for a wide list of atoms: neutral, ionized with one and two positive charges. For our case a neon neutral $a_0 = 0.933$ $a_1 = 2.353$ $a_2 = 14.871$ in atomic units.

Figure 5.18: Intensity of the 39th harmonic for different nozzle size(L) and focus position (z_0). The intensity of the laser for all cases $I=3 \times 10^{14} Wcm^{-2}$, and the confocal parameter $b = 5mm$,

The label DFT in Fig. 5.19 does reference to an potential is based on DFT [97]

$$V(r) = -\frac{1}{r}(a_0 + a_1 e^{-a_2 r} + a_3 r e^{-a_4 r} + a_5 e^{-a_6 r}); \quad (5.61)$$

$a_0 = 1.0$, $a_1 = 8.069$, $a_2 = 2.148$, $a_3 = -3.570$, $a_4 = 1.986$, $a_5 = 0.931$ and $a_6 = 0.602$ in atomic units.

Comparing the Fig. 5.19, there is not a great differences between them. The divergence angles where the intensity is different from zero are similar for each distance of z_0 , The maximum intensity is reached at similar divergence points for all focusing position z_0 .

Only there is a slightly difference for $z_0 = 1 mm$, the HF curve shows two thicker peaks than its equivalent to the DFT curves. Then, as conclusion is the atomic potential modifies slightly the intensity with the divergence.

Figure 5.19: Comparison of the intensity from the 39th harmonic with the divergence, changing the relative position (z_0) and atomic potentials. HF: denotes Hartree-Fock potential Eq. (5.60). and DFT: density functional theory Eq. (5.61).

Dependency with the pressure

The Fig. 5.20 exhibits how changes the harmonic intensity with the pressure for two $z_0 = 0, 2\text{ mm}$ points. The selection of this points is because in this range the intensity of the harmonic reaches the highest values according to the Fig. 5.17. Altucii et al [2] showed that the harmonic intensity increases only until an optimum maximum pressure is reached, beyond which it decreased.

They observed that the effect is attributed to ionization-induced defocusing of the fundamental laser beam, which reduces the peak intensity obtained in the medium and shortens the extension of the plateau the generation and propagation of high-order harmonics. At high-pressure in the range of $0.5 \sim 4\text{ atm}$ ($380 - 3040\text{ Torr}$), the ultra-short laser pulses was investigated by Nguyen Hong Shon et al[91]. The method that they used was to modify the $V_{eff}(r)$ for a single active electron generated according to the technique described in [68] [39] page 247.

The high-harmonic generation in a plasma or gas under conditions when the single-atom response is affected by neighboring ions or atoms of the medium was studied theoretically by Strelkov et al.[95] solving numerically the three dimensional Schrödinger equation for a single-electron atom in the combined fields of the neighboring particles and the laser, and average the results over different random positions of the particles using the Monte Carlo method.

This methods based on modifying the potential has not included in current

Figure 5.20: Comparison of the intensity for the 39th harmonic changing the pressure and the laser focus position (z_0).

Qhlg code.

On the other hand, the validity of the equations when the pressure increases much more than 200 *Torr* is unknown and perhaps the Lorentzian density distribution is no longer valid.

Figure 5.21: Intensity evolution over time for several pressures. $I = 3.0 \times 10^{14} \text{ W/cm}^2$, nozzle size $L = 1.2 \text{ mm}$, confocal parameter $b = 5 \text{ mm}$, laser focus at $z_0 = 0 \text{ mm}$. The ionization at end of the pulse is not high see Fig. 5.13 (a). The different behavior is because the gas is ionized. For greater pressures the number of free electrons is greater, and this electrons 'blocks' the laser propagation. This explains that at low pressure the Fig. (a) practically does not change the shape over time.

The Fig. 5.21 illustrates how some particular harmonic evolves when the pressure increases, the rest of parameters are the same for all simulations.

5.6.1 Evolution inside the interaction region.

In this section we can see what is happened in the gas neon when the laser propagates through it. For this purpose is considered the following case:

- Intensity of the laser $3.5 \times 10^{14} W/cm^2$, $\lambda = 800 \text{ nm}$
- The envelope pulse has a trapezoidal form. 2 cycles ramp-up, 10 cycles constant electric field, 2 pulses down-ramp. Total 37.359 fs .
- Confocal parameter $b = 5 \text{ mm}$.
- $z_0 = 0 \text{ mm}$ laser focus at same position of the middle of nozzle valve.
- Size of the nozzle $L = 1.8 \text{ mm}$
- Harmonic : 39^{th}
- Atom : neon, Pressure 32.0 Torr . ($1.04 \times 10^{18} \text{ atoms/cm}^3$), Distribution density : Lorentzian.

Of all the possible magnitudes that appear in the region of the interaction, only a few set of them have been selected to represent how plasma evolves over time. The magnitudes selected are intensity of the laser, modulus of the dipole moment and the ionization of gas. The dipole phase has been intentionally left outside. Although its value is crucial for all calculations, its visualization with a small number of snapshots does not give a clear picture of how this magnitude evolves.

The focusing of the laser.

The Fig. 5.22 shows how the intensity of the laser changes over time. The laser needs 5.2 fs . to reach the maximum intensity, and the same time to decrease up to the laser is disconnected. The six frames corresponds to the region where the envelope of the field is constant. The numerical scale of the intensity in all figures have to be multiplied by $\times 10^{14} W/cm^2$.

The first Fig. 5.22(a) indicates that the intensity of the laser never reaches the maximum given in the pulse characterization of $3.5 \times 10^{14} W/cm^2$. The maximum value in the frames is $3.43 \times 10^{14} W/cm^2$. The maximum value in the pulse characterization is no reached as a consequence of attenuation due to the refractive index. Over time, the maximum is reduced up to the value $3.26 \times 10^{14} W/cm^2$ in the Fig. 5.22(c).

The other effect of the refractive index over the laser field is the modification of the laser focus. Originally, when the gas is not ejected, the laser focus is placed on an axis passing through the center of the nozzle. At the beginning of

Figure 5.22: Several snapshots of the intensity laser. The key-box are in $\times 10^{14} \text{W/cm}^2$ units. The horizontal scale are in mm and it is the z axis, so the total interaction length is 1.9mm . X and Y axis are in microns. The figure evidences the evolution of the intensity as a result of the modification of the refractive index. The laser propagates from the left side to the right one in all Figs. (a)-(c). The laser propagates from left to right side.

the pulse, when the gas is present, the position of the laser focus is modified, such as the Fig. 5.22(a) shows, where the focus is moved to the left side. The same effect occurs for all frames, implying that the refractive index changes over time, and is a consequence of the appearance of free electrons because the gas is ionized. As conclusion, free electrons block the propagation of the field. This has several consequences: the cut-off frequency is reduced as a result of the reduction of the intensity, the intensity of the modulus of the dipole moment is reduced because the focus of the laser moves to other regions with lower gas density.

The modulus of dipole moment.

Figure 5.23: These pictures correspond to the polarization term given by Eq. (5.43). Then, the figure describes the evolution of the polarization over time. The figures demonstrate a strong dependence on the intensity profile

The Fig. 5.23 illustrates the local polarization given by Eq. (5.43), per gas region at the same temporal frames as Fig. 5.22. The rings of the Fig. 5.23(a) have their origin in the variation of the modulus of the dipole moment with the intensity of the laser, see Fig. 5.24.

Figure 5.24 is a cross section of Figure 5.23(a), crossing the data of Figs. 5.22(a) and 5.23(a) for the radial position to obtain dependence with the intensity. This results coincide, obviously, with the Fig. 5.8.

The ellipsoids shapes of the polarization has similar aspect to the isocurves of the intensity of the electric field, and this ellipsoids shapes coincide with the intensity isocurves over time.

Figure 5.24: Harmonic 39th. The red curve corresponds to an orthogonal slice of the figure, 5.23(a) extracted at $z = 0$. This curve is the same as Fig. 5.8 multiplied by the number of neutral atoms. The blue line is a calculation with SFA model and is similar to the Fig. 2.4 with some modifications: the curve is shifted on X-axis in 0.2 unit to the right side, because the atomic potential in Qprop generates a greater ionization potential and also on the Y-axis because the blue curve only contains the contribution of a single subcycle.

According to the specification of the simulation, the greatest number of atoms is in the middle region at $z_0 = 0$, and although the 39th harmonic is generated throughout the region, in this part of the gas is where the highest polarization values are concentrated, although the intensity decreases. See Fig. 5.23(c).

The ionization percentage of the gas.

The figure 5.25 describes the percentage of ionized atoms of each gas region. The reason for using the ionization percentage instead of ionized atoms is to avoid excessive dominance of the central region when the gas is ionized which hides the ionization of areas with smaller values.

The ionization is an accumulative process, such as was exposed in the theoretical section. The time scale is so short that the recombination of electrons is not considered and the movement of the electrons to other regions of the gas is neglected.

The ionization isocurves follow the evolution of the intensity isocurves with a temporal delay for two reasons. The first, to have an appreciable ionization the laser has to be applied for some time. The second is a consequence of the accumulation of electrons, when the focus of the laser moves to the left, the area behind the laser track has a greater number of free electrons than the area ahead. Therefore, by comparing both figures (c) of 5.25 with 5.22, the figure

Figure 5.25: Evolution of the percentage of ionization over time. The figures present a cumulative ionization because the recombination of the electrons is insignificant.

of the intensity has large values in the region along the z -axis of -0.8 mm to -0.6 mm , meanwhile, the figure of the ionization does not show large values for the same region because the intensity in this region has increased over time and reaches the highest values at the end of the constant envelope 31 fs

Chapter 6

Conclusions and future activities

To finalize this work, I will summarize the work developed in the thesis indicating what are the most important aspects.

The study of high-order harmonics began with an existing public domain code: Qprop. This code is an Schrödinger solver that permits to calculate some atomic magnitudes like the wavefunction, the dipole moment with electric field linearly and orthogonally polarized. This code has several improvements.

- The code is paralleled using OPENMP; and the efficiency of some parts in order to reduce its execution time is improved.
- The original code is a set of routines that depending on the experiment to be simulated, are selected to construct an small code that is compiled later. Qprop is modified to be a single program with an input file that allows a wide variety of cases to be executed.
- New functions to extract from its data structure the dipole moment for orthogonal lasers is added.
- New output files are created to obtain three-dimensional images.

A new program (Qhhg) is developed to study the propagation of a laser in a gas. The new program allowed us to observe some of the most interesting magnitudes such as ionization, refractive index, the phase and intensity of the harmonics in each region of the gas, as well as the variation of the laser field in the interaction region over time as a consequence of the modification of the refractive index. It also allowed us to know the angular distribution of

the intensity recorded in a detector away from the jet of gas. The code is operational but as a consequence the high computational cost required to solve the Schrödinger equation (Qprop) its development is stopped, since it did not allow to tackle two types of problems: mixing two gases and using two laser sources.

As a consequence of this, a new atomic code, the SFA-code, based on the Lewenstein model was developed to replaces Qprop. This new code is enhanced to include the saddle point approximation to improve the knowledge of the physics in the generation of harmonics. Finally, the last modification permits to use laser orthogonally polarized.

6.1 Conclusions

The main contributions of this work and the objectives achieved are summarized below.

Laser propagation in a gas medium

It wrote a completely new code to study the interactions between a jet of gas with a laser. This code reaches good agreement with the experiments. To achieve this, it is necessary to know how a laser is focused, how the index of refraction is modified with the ionization of the medium, how the atomic susceptibility is modified with the intensity of the laser. The effective sections of absorption depending on the energy of the photons is calculated by means other public code FAC.

All previous magnitudes are calculated for the central frequency of the laser and for the harmonics produced in the medium, with the end, to have a propagation model inside the code.

Interaction laser - atom

A complete new code is wrote. This code is about the laser-atom interactions in the non-relativistic regimen. This code calculates the spectrum of the harmonics, the distance of tunneling, the ionization and recombination times,

etc. With this code is published a paper based on the chapter 'Mixture gases He-Ne'. Other two papers are forecast to publish from the chapter 'Laser with orthogonal polarization'.

There is a draft of the first paper, that is based on the role of the quantum numbers $m \pm 1$ under the interaction of a laser with orthogonal polarization. The second paper, will treat about how changes the intensity of the harmonics when one of the two component of the electric field changes remaining the other fixed.

6.2 Future activities

The high-order harmonics generation with table-top lasers is a tool with an interesting future with two main research lines: the attoseconds/zeptoseconds and circularly polarized harmonics, produced with a great variety of means: diatomic gases, solids, molecules, etc. The new facilities like ELI in Hungary, and others around the world, open the possibility to deep in knowledge of the physics.

Some of improvements in the current codes are

In the SFA-code (atomic code)

- The addition to the code the calculation of the dipole acceleration instead the dipole moment to be more precisely calculations. This change will imply to develop a new transition matrix between the continuum and the ground state coupled both states by the gradient of the atomic potential.
- Include the atomic potential in calculations to treat collisional effects when the electron return to the parent ion but does not recombines.
- Improve the procedure to calculate the saddle points for two collinear electric fields when one of them is temporally delayed. This is perhaps the most complex part, the saddle point calculations with the laser pulses with variable envelope is a complex task because the algorithm is very sensible to the initial conditions to reach the final convergence.
- Include other dipolar transition matrix based on Coulomb wave-functions. One of the assumption of the SFA consists in neglect the atomic potential respect to the strength of the electric field. I think this way the new transition matrix produces similar results to the obtained with Qprop.

- Include new models for diatomic molecules. [54] [52] [63] [69] [43]
- Add to the code the angular distribution of emitted photons.

In Qhhg-code (propagation code).

- Change the Qprop code by the SFA code for the atomic calculations. This change will improve the speed of the calculations and will open the possibility of studying other experiments with orthogonally polarized lasers.
- Calculate the electric field generated by the atom based on the currents of the wavefunctions instead of the spectrum of the dipole module. This change will permit to treat with all harmonics generated by the atoms, instead of the single one in the current version. Currently the electric field generated by the atoms is calculated from the atomic spectrum selecting a single harmonic. The new procedure will calculate the full contribution from all harmonics.
- Include other laser focusing models, this will permit simulate some defects in lens or mirrors when they focus the laser.
- Perhaps the most important changes will be use a new propagation model based on Godunov scheme with Adaptive Mesh Refinement such as was described by Alfonso Barbas [11] [12], with some kind of mixture of the discrete dipole approximation Carlos Hernandez [46] [26].

Appendix A

Stokes parameters

A.1 Polarization of an Electromagnetic

The electric field $\mathbf{E}(z, t)$ of a monochromatic electric field along the z-direction with a frequency ω and an angular wave-number k can be decomposed into two orthogonal components E_x and E_y , and represented by

$$\begin{aligned}\mathbf{E}(z, t) &= \mathbf{E}_x(z, t) + \mathbf{E}_y(z, t) \\ \mathbf{E}_x(z, t) &= \mathbf{x}E_{0x}e^{i\omega t - k_z\phi_x} \\ \mathbf{E}_y(z, t) &= \mathbf{y}E_{0y}e^{i\omega t - k_z\phi_y}\end{aligned}\tag{A.1}$$

where E_{0x} and ϕ_x are the amplitude and phase, respectively, for E_x , and E_{0y} and ϕ_y are for E_y . k is wavenumber and is related to wavelength λ by $k = 2\pi/\lambda$. In vacuum, $k = \omega/c$. Let $\phi = \phi_y - \phi_x$ be the relative phase between E_y and E_x . Above equation can be simplified as

$$\mathbf{E}(z, t) = (\mathbf{x}E_{0x} + \mathbf{y}E_{0y}e^{\phi})e^{i\omega t - k_z\phi_x}\tag{A.2}$$

The projection over the plane XY at $z=0$

$$\mathbf{E}(0, t) = \mathbf{x}E_{0x}\cos(\omega t) + \mathbf{y}E_{0y}\cos(\omega t + \phi)\tag{A.3}$$

Several combination of parameters E_{0x} , E_{0y} , ϕ produce different types of polarization. For $\phi = 0$, E_{0x} and E_{0y} are in phase, then the electric field oscillates in a plane. For $\phi = \pi/2$ and $E_{0x} = E_{0y}$ the shape is circular. For else other cases, the shape is an ellipse.

Figure A.1: Figure (a). The parameters from an ellipse like minor and major axes and rotation angle Ψ , and the ellipticity angle ϵ . Figure (b), Relation between Stokes parameters and (I, ϵ, Ψ) , intensity, degree of polarization, ellipticity angle, the ellipse rotation angle.

A.2 Characteristic parameters for a polarization ellipse.

E_{0x} and E_{0y} , are the field amplitudes in the x and y directions, and ϕ is the phase retardance; a and b are the major and minor axes of the ellipse, Ψ is the orientation of the major axis with respect to the x-axis, and ϵ is the ellipticity angle which is equal to $\text{atan}(b/a)$. A polarization ellipse can be characterized by one of the following set of parameters (E_{0x}, E_{0y}, ϕ) , (a, b, Ψ) or (I, Ψ, ϵ) , where I is the intensity of the electric field.

$$\mathbf{E} = \begin{pmatrix} E_x \\ E_y \end{pmatrix} = \begin{pmatrix} E_{0x} e^{i\phi_x} \\ E_{0y} e^{i\phi_y} \end{pmatrix} \quad (\text{A.4})$$

The Stokes parameters are four intensity-based parameters used to describe the polarization state of light, represented by S_0, S_1, S_2, S_3 . The Stokes vector is the set of these Stokes parameters, defined in Cartesian coordinates $(\mathbf{x}, \mathbf{y})$ as

$$\mathbf{S} = \begin{pmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{pmatrix} = \begin{pmatrix} \langle E_x E_x^* \rangle + \langle E_y E_y^* \rangle \\ \langle E_x E_x^* \rangle - \langle E_y E_y^* \rangle \\ \langle E_x E_y^* \rangle + \langle E_y E_x^* \rangle \\ i \langle E_x E_y^* \rangle - i \langle E_y E_x^* \rangle \end{pmatrix} \quad (\text{A.5})$$

For an E field, the average bracket in Eq. A.5 represents the time average. $\langle E_x E_x^* \rangle = I_x$ is the intensity of the component of light linearly polarized in

A.2. CHARACTERISTIC PARAMETERS FOR A POLARIZATION ELLIPSE.

the x-direction. Similarly, $\langle E_y E_y^* \rangle = I_y$. All of the Stokes parameters are real numbers and are measurable.

Stokes vectors for different polarization states of light can be evaluated using Eq.A.5. The Stokes vector $\mathbf{S}$ expressed in terms of intensity I , ellipticity angle ϵ and orientation angle Ψ are , figure (b) A.1

$$\mathbf{S} = I \begin{pmatrix} 1 \\ \cos(2\epsilon)\cos(2\Psi) \\ \cos(2\epsilon)\sin(2\Psi) \\ \sin(2\epsilon) \end{pmatrix} \quad (\text{A.6})$$

Then, I, Ψ , and ϵ determine the polarization state. Stokes parameters for perfectly polarized light is given in Eq (A.5) satisfy the identity

$$S_0^2 = S_1^2 + S_2^2 + S_3^2 \quad (\text{A.7})$$

For unpolarized light with $S_1 = S_2 = S_3 = 0$ and $S = I(1, 0, 0, 0)$ Eq. A.7 does not hold. In general, light is partially polarized, i.e., part of it is perfectly polarized and the rest is unpolarized. Stokes parameters for arbitrary polarizations satisfy $S_0^2 \geq S_1^2 + S_2^2 + S_3^2$.

The degree of polarization is given by $\mathcal{P}$:

$$\mathcal{P} = \frac{\sqrt{S_1^2 + S_2^2 + S_3^2}}{S_0} \quad (\text{A.8})$$

Perfectly polarized light has $\mathcal{P} = 1$, and unpolarized light has $\mathcal{P} = 0$. Partially polarized light has $0 < \mathcal{P} < 1$. The intensity of the polarized part is $\mathcal{P}I$, and the intensity of the unpolarized part is $I(1 - \mathcal{P})$.

The degree of linear polarization $\mathcal{P}_L$ and circular polarization $\mathcal{P}_C$ are given by

$$\begin{aligned} \mathcal{P}_L &= \mathcal{P} \cos(2\epsilon) = \frac{\sqrt{S_1^2 + S_2^2}}{S_0} \\ \mathcal{P}_C &= \mathcal{P} \sin(2\epsilon) = \frac{S_3}{S_0} \end{aligned} \quad (\text{A.9})$$

The modulus of complex intensity of linear polarization in function of stokes paramaters is given by

$$L = |S_1 + iS_2| \quad (\text{A.10})$$

Other usefull parameters from the Stokes vectors are:

$$\begin{aligned}
 \Psi &= 1/2 \operatorname{atan}\left(\frac{S_2}{S_1}\right) \\
 \epsilon &= 1/2 \operatorname{atan}\left(\frac{S_3}{\sqrt{S_1^2 + S_2^2}}\right) \\
 a &= \sqrt{1/2(I\mathcal{P} + L)} \\
 b &= \sqrt{1/2(I\mathcal{P} - L)} \\
 h &= \operatorname{sign}(S_3)
 \end{aligned} \tag{A.11}$$

where a, b are the major and minor axes and h is the helicity. The helicity is the projection fo the angular momentum on the propagation direccion, it has two values -1 (LCP: Left circularly polarized) and 1 (RCP right circularly polarized).

As conclusion, for a particular hamonic $q\omega$ the projection over the axes XY of the dipole module $(D_x(q\omega), D_y(q\omega))$ is an ellipse, which can be caracterized by $|D_x(q\omega)|, |D_y(q\omega)|$ and $\phi = \phi_y - \phi_x$. Above values permit calculate the Stokes parameters and, from them, calculates minor and major axis, the helicity, the ellipticity and the ellipse rotation angle.

Appendix B

Acronyms

Mnemonic	Description
ADK	Ammosov, Delone and Krainov. Related to ionization rates
ATI	Above Threshold Ionization.
au:	atomic units.
cc	complex conjugated.
CFA	Central Field Approximation
DFT	Density Functional Theory
FAC	Flexible Atomic Code (is an atomic code)
FWHM	Full Width Half Maximum
KH	Kramers Henneberger or acceleration gauge.
KS	Kohn-Sham related with DFT.
HHG	High-order Harmonics Generation
LCP	Left Circularly Polarized.
MPI	Multiphoton Ionization.
OBI	Over the Barrier Ionization.
Qhbg	Quantum high harmonics generation (is a propagation code).
Qprop	Quantum propagation (is a atomic code).
RCP	Right Circularly Polarized.
SFA	Strong Field Approximation.
STO	Slater Type Orbitals.
TEM	Transverse Electro Magnetic
TFA	Time Frequency Analysis, also known as Gabor transformation.
TDSE	Time Dependent Schrödinger Equation
TDDFT	Time Dependent Density functional theory

Bibliography

- [1] AGOSTINI, P., AND DIMAURO, L. F. The physics of attosecond light pulses. *Reports on Progress in Physics* 67 (2004), 813. (Pages. 24 y 25.)
- [2] ALTUCCI, C., STARCZEWSKI, T., MEVEL, E., WAHLSTRÖM, C.-G., CARRÉ, B., AND L'HUILLIER, A. Influence of atomic density in high-order harmonic generation. *JOSA B* 13, 1 (Jan. 1996), 148–156. (Page. 155.)
- [3] AMMOSEV, M. V., DELONE, N., AND KRAINOV, V. P. Tunneling ionization of complex atoms and of atomic ions in an alternating electromagnetic field. *Soviet Physics JETP* 64, 6 (Dec. 1986), 1191. (Pages. 20, 59 y 142.)
- [4] ANDREI D. PLYANIN, L., AND MANZHIROV, A. V. *Handbook of Mathematics for Engineers and scientists*. Chapman and Hall/CRC, 2007. (Page. 46.)
- [5] AUGUSTE, T., CATOIRE, F., AGOSTINI, P., DIMAURO, L. F., CHIRILA, C. C., YAKOVLEV, V. S., AND SALIÈRES, P. Driving-frequency scaling of high-harmonic quantum paths. *New Journal of Physics* 14, 10 (Oct. 2012), 103014. (Page. 53.)
- [6] AUGUSTE, T., GOBERT, O., AND CARRÉ, B. Numerical study on high-order harmonic generation by a Bessel-Gauss laser beam. *Physical Review A* 78, 3 (Sept. 2008), 033411. Cited by 0012. (Pages. 121 y 133.)
- [7] AUGUSTE, T., SALIÈRES, P., WYATT, A. S., MONMAYRANT, A., WALMSLEY, I. A., CORMIER, E., ZAÏR, A., HOLLER, M., GUANDALINI, A., SCHAPPER, F., BIEGERT, J., GALLMANN, L., AND KELLER, U. Theoretical and experimental analysis of quantum path interferences in

BIBLIOGRAPHY

- high-order harmonic generation. *Physical Review A* 80, 3 (Sept. 2009), 033817. (Page. 148.)
- [8] BALCOU, P., DEDERICH, A. S., GAARDE, M. B., AND L'HUILLIER, A. Quantum-path analysis and phase matching of high-order harmonic generation and high-order frequency mixing processes in strong laser fields. *Journal of Physics B: Atomic, Molecular and Optical Physics* 32, 12 (June 1999), 2973. (Page. 24.)
- [9] BALCOU, P., AND L'HUILLIER, A. Phase-matching effects in strong-field harmonic generation. *Physical Review A* 47, 2 (1993), 1447. (Page. 133.)
- [10] BALCOU, P., SALIERES, P., L'HUILLIER, A., AND LEWENSTEIN, M. Generalized phase-matching conditions for high harmonics: The role of field-gradient forces. *Physical Review A* 55, 4 (1997), 3204. (Pages. 24 y 133.)
- [11] BARBAS, A., AND VELARDE, P. Development of a Godunov method for Maxwell's equations with Adaptive Mesh Refinement. *Journal of Computational Physics* 300 (Nov. 2015), 186–201. (Page. 166.)
- [12] BARBAS ESPA, A. *Development of AMREM a Maxwell's equations solver with Adaptive Mesh Refinement*. PhD thesis, UPM, 2015. (Page. 166.)
- [13] BAUER, D., AND KOVAL, P. QPROP: A Schrodinger-solver for intense laser-atom interaction, July 2005. (Pages. 66 y 132.)
- [14] BAUER, D., AND KOVAL, P. Qprop: A Schrödinger-solver for intense laser-atom interaction. *Computer physics communications* 174, 5 (2006), 396–421. (Pages. 71, 78, 88 y 97.)
- [15] BAUER, D., MILOSEVIC, D. B., AND BECKER, W. Strong-field approximation for intense-laser-atom processes: The choice of gauge. *Physical Review A* 72, 2 (2005), 023415. (Pages. 25, 72 y 74.)
- [16] BAUER, D., AND MULSER, P. Exact field ionization rates in the barrier-suppression regime from numerical time-dependent Schrödinger-equation calculations. *Physical Review A* 59, 1 (1999), 569. (Pages. 60 y 142.)
- [17] BAUER, J. Coulomb-corrected Volkov-type solution for an electron in an intense circularly polarized laser field. *Journal of Physics B: Atomic, Molecular and Optical Physics* 34, 7 (Apr. 2001), 1343–1347. (Page. 30.)

- [18] BECKER, W., CHICHKOV, B. N., AND WELLEGEHAUSEN, B. Schemes for the generation of circularly polarized high-order harmonics by two-color mixing. *Physical Review A* 60, 2 (Aug. 1999), 1721–1722. (Page. 87.)
- [19] BECKER, W., LOHR, A., KLEBER, M., AND LEWENSTEIN, M. A unified theory of high-harmonic generation: Application to polarization properties of the harmonics. *Physical Review A* 56, 1 (July 1997), 645–656. (Pages. 25 y 88.)
- [20] BECKER, W., LONG, S., AND MCIVER, J. K. Modeling harmonic generation by a zero-range potential. *Physical Review A* 50, 2 (Aug. 1994), 1540–1560. (Page. 88.)
- [21] BERRY, M. V. Stokes’ phenomenon; smoothing a victorian discontinuity. *Publications Mathématiques de l’IHÉS* 68 (1988), 211–221. (Page. 54.)
- [22] BERRY, M. V. Uniform asymptotic smoothing of Stokes’s discontinuities. *Proc. R. Soc. A* 422 (1989), 7–21. (Page. 54.)
- [23] BLEISTEIN, N., AND HANDELSMAN, R. A. *Asymptotic Expansion of Integrals*, 1975 ed. Dover Publications Inc., 2015. (Page. 55.)
- [24] BOYD, R. W. *Nonlinear optics*. Academic press, 2003. (Pages. 143 y 146.)
- [25] BÉJOT, P., CORMIER, E., HERTZ, E., LAVOREL, B., KASPARIAN, J., WOLF, J.-P., AND FAUCHER, O. High-Field Quantum Calculation Reveals Time-Dependent Negative Kerr Contribution. *Physical Review Letters* 110, 4 (Jan. 2013). (Page. 146.)
- [26] CARLOS, H. G. *Coherent attosecond light sources based on high-order harmonic generation: influence of the propagation effects*. PhD thesis, Universidad de Salamanca, Jan. 2013. (Page. 166.)
- [27] CHANG, Z. *Fundamentals of Attosecond Optics*, 1 edition ed. CRC Press, Boca Raton, Feb. 2011. (Page. 44.)
- [28] CHANG, Z., RUNDQUIST, A., WANG, H., MURNANE, M. M., AND KAPTEYN, H. C. Generation of Coherent Soft X Rays at 2.7 nm Using High Harmonics. *Physical Review Letters* 79, 16 (Oct. 1997), 2967–2970. (Page. 23.)

BIBLIOGRAPHY

- [29] CHEN, M., CORMIER-MICHEL, E., GEDDES, C. G. R., BRUHWILER, D. L., YU, L. L., ESAREY, E., SCHROEDER, C. B., AND LEEMANS, W. P. Numerical modeling of laser tunneling ionization in explicit particle-in-cell codes. *Journal of Computational Physics* 236 (Mar. 2013), 220–228. (Page. 60.)
- [30] CHIRILĂ, C. C., DREISSIGACKER, I., VAN DER ZWAN, E. V., AND LEIN, M. Emission times in high-order harmonic generation. *Physical Review A* 81, 3 (Mar. 2010), 033412. (Page. 66.)
- [31] CORKUM, P. B. Plasma perspective on strong field multiphoton ionization. *Physical Review Letters* 71, 13 (1993), 1994–1997. (Pages. 22 y 23.)
- [32] CORSI, C., PIRRI, A., SALI, E., TORTORA, A., AND BELLINI, M. Direct Interferometric Measurement of the Atomic Dipole Phase in High-Order Harmonic Generation. *Physical Review Letters* 97, 2 (July 2006), 023901. (Page. 49.)
- [33] COWAN, R. D. *The Theory of Atomic Structure and Spectra*. University of California Press, Berkeley, Sept. 1981. (Page. 43.)
- [34] EICHMANN, H., EGBERT, A., NOLTE, S., MOMMA, C., WELLEGEHAUSEN, B., BECKER, W., LONG, S., AND MCIVER, J. K. Polarization-dependent high-order two-color mixing. *Physical Review A* 51, 5 (May 1995), R3414–R3417. (Page. 87.)
- [35] FERRÉ, A., HANDSCHIN, C., DUMERGUE, M., BURG, F., COMBY, A., DESCAMPS, D., FABRE, B., GARCIA, G. A., GÉNEAUX, R., MERCERON, L., MÉVEL, E., NAHON, L., PETIT, S., PONS, B., STAEDTER, D., WEBER, S., RUCHON, T., BLANCHET, V., AND MAIRESSE, Y. A table-top ultrashort light source in the extreme ultraviolet for circular dichroism experiments. *Nature Photonics* 9, 2 (Dec. 2014), 93–98. (Page. 87.)
- [36] FIGUEIRA DE MORISSON FARIA, C., SCHOMERUS, H., AND BECKER, W. High-order above-threshold ionization: The uniform approximation and the effect of the binding potential. *Physical Review A* 66, 4 (Oct. 2002), 043413. (Page. 55.)

- [37] GAARDE, M. B., SALIN, F., CONSTANT, E., BALCOU, P., SCHAFER, K. J., KULANDER, K. C., AND L'HUILLIER, A. Spatiotemporal separation of high harmonic radiation into two quantum path components. *Physical Review A* 59, 2 (Feb. 1999), 1367–1373. (Page. 148.)
- [38] GAARDE, M. B., TATE, J. L., AND SCHAFER, K. J. Macroscopic aspects of attosecond pulse generation. *Journal of Physics B: Atomic, Molecular and Optical Physics* 41, 13 (July 2008), 132001. (Page. 130.)
- [39] GAVRILIA, M. *Atoms in intense laser fields*. Academic Press, 1992. (Pages. 130, 136 y 155.)
- [40] GONZÁLEZ-FERNÁNDEZ, A., AND VELARDE, P. Quantum interference of high-order harmonics from mixed gases. *Physical Review A* 94, 2 (Aug. 2016), 023804. (Pages. 10, 92, 101 y 109.)
- [41] GRIBAKIN, G. F., AND KUCHIEV, M. Y. Multiphoton detachment of electrons from negative ions. *Physical Review A* 55, 5 (May 1997), 3760–3771. (Pages. 45, 46, 51, 71 y 73.)
- [42] GU, M. F. The flexible atomic code. *Canadian Journal of Physics* 86, 5 (May 2008), 675–689. (Pages. 13, 139 y 140.)
- [43] GUO, J., GE, X.-L., ZHONG, H., ZHAO, X., ZHANG, M., JIANG, Y., AND LIU, X.-S. Influence of vibrational states on high-order-harmonic generation and an isolated attosecond pulse from a $\{\mathrm{N}\}_2$ molecule. *Physical Review A* 90, 5 (Nov. 2014), 053410. (Page. 166.)
- [44] HANS A. BETHE, AND EDWIN E. SALPETER. *Quantum Mechanics of One and Two-Electron Atoms*. Springer-Verlag, 1957. (Pages. 45, 46, 71, 73, 74 y 89.)
- [45] HERNÁNDEZ-GARCÍA, C., PÉREZ-HERNÁNDEZ, J. A., POPMINTCHEV, T., MURNANE, M. M., KAPTEYN, H. C., JARON-BECKER, A., BECKER, A., AND PLAJA, L. Zeptosecond High Harmonic keV X-Ray Waveforms Driven by Midinfrared Laser Pulses. *Physical Review Letters* 111, 3 (July 2013), 033002. (Page. 67.)
- [46] HERNÁNDEZ-GARCÍA, C., PÉREZ-HERNÁNDEZ, J. A., RAMOS, J., JARQUE, E. C., ROSO, L., AND PLAJA, L. High-order harmonic propa-

BIBLIOGRAPHY

- gation in gases within the discrete dipole approximation. *Physical Review A* 82, 3 (Sept. 2010), 033432. (Page. 166.)
- [47] HOHENBERG, P., AND KOHN, W. Inhomogeneous Electron Gas. *Physical Review* 136, 3B (Nov. 1964), B864–B871. (Page. 60.)
- [48] IVANOV, I. A., AND KHEIFETS, A. S. On the use of the Kramers–Henneberger Hamiltonian in multi-photon ionization calculations. *Journal of Physics B: Atomic, Molecular and Optical Physics* 38, 13 (2005), 2245. (Page. 30.)
- [49] JACKSON, J. D. *Classical Electrodynamics Third Edition*, 3 edition ed. Wiley, New York, Aug. 1998. (Page. 135.)
- [50] JAEGLE, P. *Coherent Sources of XUV Radiation*. Springer Science+Business Media, Inc, [New York], 2006. (Page. 141.)
- [51] JOACHAIN, C. J., KYLSTRA, N. J., AND POTVLIEGE, R. M. *Atoms in Intense Laser Fields*. Cambridge University Press, Dec. 2011. (Pages. 31, 42, 45, 52 y 57.)
- [52] KANAI, T., MINEMOTO, S., AND SAKAI, H. Ellipticity Dependence of High-Order Harmonic Generation from Aligned Molecules. *Physical Review Letters* 98, 5 (Feb. 2007), 053002. (Page. 166.)
- [53] KANAI, T., TAKAHASHI, E. J., NABEKAWA, Y., AND MIDORIKAWA, K. Destructive Interference during High Harmonic Generation in Mixed Gases. *Physical Review Letters* 98, 15 (Apr. 2007), 153904. (Pages. 8, 72, 75 y 77.)
- [54] KANAI, T., TAKAHASHI, E. J., NABEKAWA, Y., AND MIDORIKAWA, K. Observing molecular structures by using high-order harmonic generation in mixed gases. *Physical Review A* 77, 4 (Apr. 2008), 041402. (Page. 166.)
- [55] KELDYSH, L. V. Ionization in the field of strong electromagnetic wave. *Soviet Physics JETP* 20, 5 (May 1965), 1307. (Page. 21.)
- [56] KFIR, O., GRYCHTOL, P., TURGUT, E., KNUT, R., ZUSIN, D., POPMINTCHEV, D., POPMINTCHEV, T., NEMBACH, H., SHAW, J. M., FLEISCHER, A., KAPTEYN, H., MURNANE, M., AND COHEN, O. Generation of bright phase-matched circularly-polarized extreme ultraviolet high harmonics. *Nature Photonics* 9, 2 (Dec. 2014), 99–105. (Page. 87.)

- [57] KOHLER, M. C., OTT, C., RAITH, P., HECK, R., SCHLEGEL, I., KEITEL, C. H., AND PFEIFER, T. High Harmonic Generation Via Continuum Wave-Packet Interference. *Physical Review Letters* 105, 20 (Nov. 2010), 203902. (Page. 26.)
- [58] KOVÁCS, K., AND TOŞA, V. Quantum trajectories of electrons in arbitrary laser fields. *Journal of Modern Optics* 57, 11 (June 2010), 977–983. (Page. 47.)
- [59] KRAUSE, J. L., SCHAFER, K. J., AND KULANDER, K. C. High-order harmonic generation from atoms and ions in the high intensity regime. *Physical Review Letters* 68, 24 (June 1992), 3535–3538. (Page. 23.)
- [60] KULANDER, K. C., SCHAFER, K. J., AND KRAUSE, J. L. Super-Intense Laser-Atom Physics. *NATO Advanced Studies Institute* 316 (1993), 95–110. (Page. 22.)
- [61] LAGO, A., HILBER, G., AND WALLENSTEIN, R. Optical-frequency conversion in gaseous media. *Physical Review A* 36, 8 (1987), 3827. (Page. 130.)
- [62] LAMBERT, G., VODUNGBO, B., GAUTIER, J., MAHIEU, B., MALKA, V., SEBBAN, S., ZEITOUN, P., LUNING, J., PERRON, J., ANDREEV, A., STREMOUKHOV, S., ARDANA-LAMAS, F., DAX, A., HAURI, C. P., SARDINHA, A., AND FAJARDO, M. Towards enabling femtosecond helicity-dependent spectroscopy with high-harmonic sources. *Nature Communications* 6 (Feb. 2015), 6167. (Page. 87.)
- [63] LE, A.-T., MORISHITA, T., LUCCHESI, R. R., AND LIN, C. D. Theory of High Harmonic Generation for Probing Time-Resolved Large-Amplitude Molecular Vibrations with Ultrashort Intense Lasers. *Physical Review Letters* 109, 20 (Nov. 2012), 203004. (Page. 166.)
- [64] LEWENSTEIN, M., BALCOU, P., IVANOV, M. Y., L’HUIILLIER, A., AND CORKUM, P. B. Theory of high-harmonic generation by low-frequency laser fields. *Physical Review A* 49, 3 (Mar. 1994), 2117–2132. (Pages. 24, 25, 44, 45, 47, 71, 73, 88 y 92.)
- [65] LEWENSTEIN, M., SALIERES, P., AND L’HUIILLIER, A. Phase of the atomic polarization in high-order harmonic generation. *Physical Review A* 52, 6 (Dec. 1995), 4747–4754. (Page. 88.)

BIBLIOGRAPHY

- [66] L'HUILLIER, A., SCHAFER, K. J., AND KULANDER, K. C. Theoretical aspects of intense field harmonic generation. *Journal of Physics B: Atomic, Molecular and Optical Physics* 24, 15 (Aug. 1991), 3315–3341. (Page. 136.)
- [67] LUCCHINI, M., CALEGARI, F., KIM, K., SANSONE, G., AND NISOLI, M. Nonadiabatic quantum path analysis of the high-order harmonic generation in a highly ionized medium. *New Journal of Physics* 14, 3 (Mar. 2012), 033009. (Page. 60.)
- [68] L'HUILLIER, A., BALCOU, P., CANDEL, S., SCHAFER, K. J., AND KULANDER, K. C. Calculations of high-order harmonic-generation processes in xenon at 1064 nm. *Physical Review A* 46, 5 (Sept. 1992), 2778–2790. (Page. 155.)
- [69] MCFARLAND, D. K. *HHG from multiple orbital in molecular Nitrogen*. PhD thesis, Stanford University, Mar. 2010. (Page. 166.)
- [70] MILOSEVIC, D. B., AND BECKER, W. Attosecond pulse trains with unusual nonlinear polarization. *Physical Review A* 62, 1 (June 2000), 011403. (Page. 88.)
- [71] MILOSEVIC, D. B., AND BECKER, W. Role of long quantum orbits in high-order harmonic generation. *Physical Review A* 66, 6 (Dec. 2002), 063417. (Pages. 56 y 57.)
- [72] MILOSEVIC, D. B., BECKER, W., AND KOPOLD, R. Generation of circularly polarized high-order harmonics by two-color coplanar field mixing. *Physical Review A* 61, 6 (May 2000), 063403. (Page. 88.)
- [73] MILOSEVIC, D. B., PAULUS, G. G., BAUER, D., AND BECKER, W. Above-threshold ionization by few-cycle pulses. *Journal of Physics B: Atomic, Molecular and Optical Physics* 39, 14 (July 2006), R203–R262. (Page. 52.)
- [74] MULLER, H. G. An efficient propagation scheme for the time-dependent Schrödinger equation in the velocity gauge. *Laser Physics* 9 (1999), 138–148. (Pages. 63 y 66.)

- [75] MURAKAMI, M., KOROBKIN, O., AND HORBATSCH, M. High-harmonic generation from hydrogen atoms driven by two-color mutually orthogonal laser fields. *Physical Review A* 88, 6 (Dec. 2013), 063419. (Page. 88.)
- [76] OLGA SMIRNOVA. *Multielectron High Harmonic Generation: simple man on complex plane*. (Pages. 51 y 53.)
- [77] PARR, R. G., AND WEITAO, Y. *Density-Functional Theory of Atoms and Molecules*. Oxford University Press, May 1994. Google-Books-ID: mGOpScSIwU4C. (Page. 60.)
- [78] POPMINTCHEV, T., CHEN, M.-C., ARPIN, P., MURNANE, M. M., AND KAPTEYN, H. C. The attosecond nonlinear optics of bright coherent X-ray generation. *Nature Photonics* 4, 12 (Dec. 2010), 822–832. (Page. 133.)
- [79] POPMINTCHEV, T., CHEN, M.-C., BAHABAD, A., GERRITY, M., SIDORENKO, P., COHEN, O., CHRISTOV, I. P., MURNANE, M. M., AND KAPTEYN, H. C. Phase matching of high harmonic generation in the soft and hard X-ray regions of the spectrum. *Proceedings of the National Academy of Sciences* 106, 26 (June 2009), 10516–10521. (Page. 133.)
- [80] PRIORI, E., CERULLO, G., NISOLI, M., STAGIRA, S., DE SILVESTRI, S., VILLORESI, P., POLETO, L., CECCHERINI, P., ALTUCCI, C., BRUZZESE, R., AND DE LISIO, C. Nonadiabatic three-dimensional model of high-order harmonic generation in the few-optical-cycle regime. *Physical Review A* 61, 6 (May 2000), 063801. (Pages. 32 y 148.)
- [81] REED, V. C., AND BURNETT, K. Ionization of atoms in intense laser pulses using the Kramers-Henneberger transformation. *Physical Review A* 42 (1990), 3152–3155. (Page. 30.)
- [82] RUIZ, C., HOFFMANN, D. J., TORRES, R., CHIPPERFIELD, L. E., AND MARANGOS, J. P. Control of the polarization of attosecond pulses using a two-color field. *New Journal of Physics* 11, 11 (Nov. 2009), 113045. (Page. 88.)
- [83] RÍO, C. S. D. *Introducción a la teoría del átomo*. Alhambra, 1977. (Page. 153.)

BIBLIOGRAPHY

- [84] SALIERES, P., L'HUILLIER, A., AND LEWENSTEIN, M. Coherence Control of High-Order Harmonics. *Physical Review Letters* 74, 19 (May 1995), 3776–3779. (Pages. 14, 148, 149, 150 y 151.)
- [85] SALIÈRES, P., CARRÉ, B., DÉROFF, L. L., GRASBON, F., PAULUS, G. G., WALTHER, H., KOPOLD, R., BECKER, W., MILOSEVIC, D. B., SANPERA, A., AND LEWENSTEIN, M. Feynman's Path-Integral Approach for Intense-Laser-Atom Interactions. *Science* 292, 5518 (May 2001), 902–905. (Pages. 33 y 88.)
- [86] SANSONE, G. Quantum path analysis of isolated attosecond pulse generation by polarization gating. *Physical Review A* 79, 5 (May 2009), 053410. (Pages. 56, 57 y 88.)
- [87] SANSONE, G., VOZZI, C., STAGIRA, S., AND NISOLI, M. Nonadiabatic quantum path analysis of high-order harmonic generation: Role of the carrier-envelope phase on short and long paths. *Physical Review A* 70, 1 (July 2004), 013411. (Pages. 26 y 81.)
- [88] SCHAFFER, K. J., GAARDE, M. B., HEINRICH, A., BIEGERT, J., AND KELLER, U. Strong Field Quantum Path Control Using Attosecond Pulse Trains. *Physical Review Letters* 92, 2 (Jan. 2004), 023003. (Page. 67.)
- [89] SCHIESSL, K., ISHIKAWA, K. L., PERSSON, E., AND BURGDÖRFER, J. Quantum Path Interference in the Wavelength Dependence of High-Harmonic Generation. *Physical Review Letters* 99, 25 (Dec. 2007), 253903. (Page. 54.)
- [90] SCHLEICH, W. P. E: Airy Function and Stokes phenomenon. In *Quantum Optics in Phase Space*. Wiley-VCH Verlag GmbH & Co. KGaA, 2001, pp. 621–627. (Page. 54.)
- [91] SHON, N. H., SUDA, A., AND MIDORIKAWA, K. Generation and propagation of high-order harmonics in high-pressure gases. *Physical Review A* 62, 2 (July 2000), 023801. (Page. 155.)
- [92] SMIRNOVA, O. V. Validity of the Kramers-Henneberger approximation. *Journal of Experimental and Theoretical Physics* 90, 4 (2000), 609–616. (Page. 30.)

- [93] STRELKOV, V. V. Theory of high-order harmonic generation and attosecond pulse emission by a low-frequency elliptically polarized laser field. *Physical Review A* 74, 1 (July 2006), 013405. (Pages. 26 y 88.)
- [94] STRELKOV, V. V., KHOKHLOVA, M. A., GONOSKOV, A. A., GONOSKOV, I. A., AND RYABIKIN, M. Y. High-order harmonic generation by atoms in an elliptically polarized laser field: Harmonic polarization properties and laser threshold ellipticity. *Physical Review A* 86, 1 (July 2012), 013404. (Page. 88.)
- [95] STRELKOV, V. V., PLATONENKO, V. T., AND BECKER, A. High-harmonic generation in a dense medium. *Physical Review A* 71, 5 (May 2005), 053808. (Page. 155.)
- [96] TONG, X.-M., AND CHU, S.-I. Generation of circularly polarized multiple high-order harmonic emission from two-color crossed laser beams. *Physical Review A* 58, 4 (Oct. 1998), R2656–R2659. (Page. 87.)
- [97] TONG, X. M., AND LIN, C. D. Empirical formula for static field ionization rates of atoms and molecules by lasers in the barrier-suppression regime. *Journal of Physics B: Atomic, Molecular and Optical Physics* 38, 15 (Aug. 2005), 2593–2600. (Pages. 60 y 154.)
- [98] VOZZI, C., NEGRO, M., CALEGARI, F., STAGIRA, S., KOVÁCS, K., AND TOSA, V. Phase-matching effects in the generation of high-energy photons by mid-infrared few-cycle laser pulses. *New Journal of Physics* 13, 7 (2011), 073003. (Page. 133.)
- [99] WINFUL, H. G. Physical origin of the Gouy phase shift. *Optics letters* 26, 8 (2001), 485. (Page. 134.)
- [100] WONG, R. *Asymptotic Approximation of Integrals*. SIAM: Society for Industrial and Applied Mathematics, Philadelphia, Aug. 2001. (Page. 55.)
- [101] YAN, T.-M., POPRUZHENKO, S. V., VRAKING, M. J. J., AND BAUER, D. Low-Energy Structures in Strong Field Ionization Revealed by Quantum Orbits. *Physical Review Letters* 105, 25 (Dec. 2010), 253002. (Page. 59.)
- [102] YAN TIAN-MIN. *Trajectory-Based Coulomb-Corrected Strong Field Approximation*. PhD thesis, 2013. (Page. 59.)

BIBLIOGRAPHY

- [103] ZAIR, A., HOLLER, M., GUANDALINI, A., SCHAPPER, F., BIEGERT, J., GALLMANN, L., KELLER, U., WYATT, A. S., MONMAYRANT, A., WALMSLEY, I. A., CORMIER, E., AUGUSTE, T., CAUMES, J. P., AND SALIÈRES, P. Quantum Path Interferences in High-Order Harmonic Generation. *Physical Review Letters* 100, 14 (Apr. 2008), 143902. (Page. 148.)

List of publications and international conferences

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- COST MP1203: Ultrafast Science and Technology Spain 2015: Intensity and wave-front produced by HHG. Dubrovnik, Croatia (2014). Participatory - poster
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in time-dependent simulations of low-Z materials illuminated by a high-intensity X-ray laser”, High Energy Density Physics 9 (2013) 542-547.

- *François de Dortran; Dominique Kilbane; J Vyskoeil; Philippe Zeitoun; Agustín González Fernández; Alberto García de la Varga; Olivier Guibaud; David Portillo García; Alfonso Bargas Espá; Pedro Velarde Mayol.* "Atomic and plasma physics software and satabases for the simulation of short wavelength sources. Short Wavelength Laboratory Sources, Principles and Practices." pp. 3 - 13. Royal Society of Chemistry, 2015.
- *Alberto García de la Varga; François de Gaufridy; Pedro Velarde Mayol; Manuel Cotelo Ferreira; David Portillo García; Alfonso Barbas Espá; Agustín González Fernández; Philippe Zeitoun.* "Influence of non-Maxwellian electron distribution in low-Z elements iluminated by a high intensity x-ray lasers". Nonlinear Optics and Applications VII. 8772, SPIE, 08/05/2013.

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