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TIAGO GUALBERTO BEZERRA DE SOUZA

Characterization of gases nonlinear refractive index and energetic deep UV
generation in hollow core fiber

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Characterization of gases nonlinear refractive index and energetic deep UV
generation in hollow core fiber

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Physics at the Instituto de Física de São
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“The task is not so much to see what no one has seen yet;
but to think what nobody has thought yet,
about what everybody sees.”

Arthur Schopenhauer

ABSTRACT

SOUZA, T. G. B. **Characterization of gases nonlinear refractive index and energetic deep UV generation in hollow core fiber.** 2019. 96 p. Thesis (Doctor of Science) - Instituto de Física de São Carlos, Universidade de São Paulo, São Carlos, 2019.

We have measured, for the first time, the magnitude and temporal pulse width dependency of the nonlinear refractive index of gaseous samples using the nonlinear ellipse rotation effect. Due to their low density, gases nonlinear refractive index are hardly accessed through available techniques commonly used for solid and liquid samples. There are techniques based on spectral broadening, interferometry, four wave-mixing, and filamentation which probe gases nonlinear refractive index, though each one present some sort of limitation or constrain. In order to provide an alternative to the previous techniques, we present a novel experimental setup to measure gases nonlinear refractive index for several excitation pulse width. The weak gases nonlinearity was overcome by extending the gas-light interaction length using a hollow core fiber, also known as capillary. By this way, we measured the nonlinear refractive index of helium, argon, nitrogen, and oxygen, along with their mixture into the atmospheric gas. Molecular gases nonlinear refractive index contribution results from its intra-pulse molecular alignment, which is dependent on the pulse width. On the other hand, we have profited from noble gases instantaneous electronic contribution, as they are monoatomic gases, to obtain a temporal pulse calibration based on argon nonlinear ellipse rotation signal. In order to unveil the transition and the amount of each contribution in gases, we have studied and characterized the effective nonlinear refractive index of the studied samples varying the pulse width from 40 fs up to 3 ps in a continuous manner. An empirical model has been proposed in order to distinguish the electronic nonresonant from the molecular orientation contribution for the effective nonlinear refractive index. In the second part of this work, we have explored infrared pulses spectrum broadening in hollow core fiber filled with noble gases, followed by proper temporal compression to reach sub-10 fs IR pulse, which was used to excite a soliton breaking phenomena in order to study the generation of energetic and ultrashort tunable deep ultraviolet light source. The dispersion wave emission in gas filled hollow core fibers have been recently explored for this very end. New sources of light, with enough energy and ultrashort pulse width, capable to excite nonlinearities in the materials, has been extremely desired for spectroscopy in the deep UV region.

Keywords: Nonlinear ellipse rotation. Nonlinear refractive index. Atmospheric gases. UV generation. Ultrafast optics.

RESUMO

SOUZA, T. G. B. **Caracterização do índice de refração não linear em gases e geração de UV profundo e energético em fibras ocas.** 2019. 96 p. Tese (Doutorado em Ciências) - Instituto de Física de São Carlos, Universidade de São Paulo, São Carlos, 2019.

Nós medimos, pela primeira vez, a magnitude e a dependência com a duração temporal do feixe de excitação do índice de refração não linear de gases utilizando a rotação não linear da polarização elíptica. O índice de refração não linear de amostras gasosas é dificilmente acessível através das técnicas experimentais utilizadas usualmente em sólidos e líquidos. Técnicas baseadas no alargamento espectral, interferometria, mistura de quatro ondas, e filamentação medem o índice de refração não linear de gases, porém cada uma delas apresenta limitações e características distintas. Afim de sobrepujar essas limitações, exploramos um novo método para medir o índice de refração não linear em gases de uma forma ainda mais sensível e para várias durações de pulso. A fraca magnitude dos efeitos não lineares em gases foi superada estendendo o comprimento de interação confinando os gases em fibras oca. Dessa forma, medimos o índice de refração não linear de amostras de hélio, argônio, nitrogênio, oxigênio, além da composição destes contida no ar atmosférico. Nós demonstramos como a contribuição do alinhamento intra-pulso das moléculas que compõem os gases moleculares para o valor do índice de refração não linear dependem da duração do pulso. Exploramos a contribuição instantânea dos gases nobres, para medir o valor da duração do pulso através do sinal de RNLPE medido no argônio. Assim, para quantificar e distinguir a quantidade de cada contribuição nos gases, caracterizamos o valor efetivo do índice de refração não linear das amostras estudadas variando a duração temporal continuamente desde 40 fs até 3 ps. Propusermos um modelo empírico para avaliar a contribuição oriunda de efeitos puramente eletrônicos e de origem em orientação molecular para o valor efetivo do índice de refração não linear. Na segunda etapa deste trabalho, exploramos o alargamento espectral em fibra oca preenchidos com gases nobres, seguido de compressão temporal para produzir pulsos sub 10 fs no infra-vermelho, que por sua vez foram utilizados para induzir a fissão em sólitons ópticos para estudar a geração de ultra-violeta profundo, sintonizável, energético e ultracurto. O fenômeno de emissão de onda dispersiva em fibras ocas preenchidas com gases tem sido recentemente explorado para esse fim. Novas fontes de luz sintonizáveis na região do UV profundo, com intensidade suficiente para induzir fenômenos não lineares, desejada para espectroscopia de materiais.

Palavras-chave: Rotação não linear da polarização elíptica. Índice de refração não linear. Gases atmosféricos. Geração de UV. Óptica ultrarrápida.

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LIST OF ABBREVIATIONS AND ACRONYMS

AC	Auto-Correlation
Air	Atmospheric Air
Ar	Argon
BBO	Beta Barium Borate
CFWM	Chirped Four Wave Mixing
CM	Chirped Mirror
CPA	Chirped Pulse Amplification
DFG	Difference-Frequency Generation
DFWM	Degenerate Four Wave Mixing
DUV	Deep Ultra-Violet
DWE	Dispersive Wave Emission
ED	External Diameter
ENR	Electronic Nonresonant
FROG	Frequency Resolved Optical Gating
FTL	Fourier-Transform Limit
FWM	Four Wave Mixing
GVD	Group Velocity Dispersion
HCF	Hollow Core Fiber
He	Helium
ICFO	The Institute of Photonics Sciences
ID	Inner Diameter
IR	Infra-Red
Kr	Krypton
KrF	Krypton Fluoride
MO	Molecular Orientation
MS	Marcatili-Schmeltzer
N ₂	Molecular Nitrogen
Ne	Neon
NER	Nonlinear Ellipse Rotation
NLO	Nonlinear Optical
O ₂	Molecular Oxygen

OPO	Optical Parametric Oscillation
P	Prism
PCF	Photonic Crystal Fiber
PM	Plane Mirror
S	Stopper
SD	Self-Diffraction
SFG	Sum-Frequency Generation
SHG	Second Harmonic Generation
SIF	Step-Index Fiber
SP	Spectrum Photometer
SPM	Self-Phase Modulation
THG	Third-Harmonic Generation
TOD	Third-Order Dispersion
UV	Ultraviolet
Xe	Xenon
XPM	Cross Phase Modulation

LIST OF SYMBOLS

a	Core Radius
A	Frequency Dependent Electric Field Amplitude
A_{eff}	Effective Mode Area
B	Magnetic Flux Density
B_{eff}	Effective B
B_{ENR}	Electronic Nonresonant Value of B
B_{MO}	Molecular Orientation Value of B
c	Vacuum Speed of Light
C	Rotation Symmetry Operation
CaF_2	Calcium Fluoride
CS_2	Carbon Disulfide
D	Electric Flux Density
d_0	Reference Gratings Distance
E	Electric Field
$\tilde{E}$	Fourier Transformed Electric Field
$\bar{E}_p$	Average Pulse Energy
E_z	Longitudinal Electric Field
EH_{mn}	Hybrid Electric Mode
f	Focus Length
F	Radial Dependent Electric Field Amplitude
H	Magnetic Field
H_z	Longitudinal Magnetic Field.
HE_{mn}	Hybrid Magnetic Mode
I	Light Intensity
J	Electric Current Density
J_m	Bessel Function
k_0	Vacuum Wave Number
K_m	Neuman Function
L	Fiber Length
L_D	Dispersion Length
L_{NL}	Nonlinear Length

M	Magnetic Polarization Density
MgF_2	Magnesium Fluoride
n	Index of Refraction
N	Density of Particles
$n_{2,eff}$	Effective Nonlinear Refractive Index
$n_{2,ENR}$	Electronic Nonresonant Value of n_2
$n_{2,MO}$	Molecular Orientation Value of n_2
n_2	Nonlinear Index of Refraction
n_c	Core Refractive Index
n_d	Cladding Refractive Index
N_m	Number of Mode
N_s	Soliton-order
p	Pressure
P	Electric Polarization Density
$P^{(2)}$	Second-Order Polarization Density
$P^{(3)}$	Third-Order Polarization Density
$P^{(4)}$	Fourth-Order Polarization Density
$P^{(6)}$	Sixth-Order Polarization Density
P^L	Linear Polarization Density
P^{NL}	Nonlinear Polarization Density
PW	Average Power
PW_0	Peak Power
R_{ij}	Rotation Matrix Operator
R_r	Repetition Rate
S_i	Inversion Symmetry Operator
sgn	Signal
SiO_2	Silicon Dioxide
$SVEA$	Slowly Variation Envelope Amplitude
T	Temperature
t_g	Growth Parameter
t_{gv}	Group Velocity Propagation Time Frame
t_N	Normalized Time Frame

U	Normilized Electric Field Amplitude
u_{mn}	Bessel root
V	V-mode Number
v_g	Group Velocity
V_U	Normilized Electric Field Amplitude
w_0	Beam Waist
z	Longitudinal Coordinate
z_R	Rayleigh Range
α	Absorption Coefficient
β	Propagation Wave Number
β_0	Zero-Order Wave Number
β_1	First-Order Wave Number
β_2	Second-Order Wave Number
β_3	Third-Order Wave Number
β_{eff}	Effective Wave Number
γ	Nonlinear Parameter
δ	Dirac Delta Function
Δ	Normilized Refractive Index Difference
δ_{ijk}	Kronecker's Delta
Δd	Gratings Relative Distance
δn	Refractive Index Variation
Δn	Circular Birefringence
$\delta \alpha$	Absorption coefficient Variation
$\Delta \theta$	Ellipse Rotation Angle
$\Delta \Phi$	Cross Phase Shift
$\Delta \Psi$	Relative Modulation Phase Shift
$\delta \omega$	Frequency Variation
ε	Electric Permittivity
ε_0	Vaccum Permittivity
κ	Core Transverse Wave Number
λ_0	Vacuum Wavelength
λ_{zd}	Second-Order Zero Dispersion Wavelength
μ_0	Vaccum Permeability

ρ	Radial Coordinate
ρ_f	Electric Charge Density
σ	Cladding Transverse Wave Number
σ_{ij}	Mirror Symmetry Operation
τ_p	Temporal Pulse Width
φ	Ellipese Eccentricity
ϕ	Angular Coordinate
$\chi^{(1)}$	First-Order Electric Susceptibility
$\chi^{(3)}$	Third-Order Electric Susceptibility
Ψ_0	Reference Modulation Phase Shift
ψ_{NL}	Nonlinear Phase
ω	Light Frequency
ω_0	Central Frequency

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1 INTRODUCTION

Whenever high intensity pulses propagate through a gaseous media, it gives rise to nonlinear optical (NLO) effects that may alter their spatial, spectral and temporal characteristics.¹ The knowledge of NLO effects on gaseous media is fundamental to applications such as high harmonic generation², attosecond physics³⁻⁵, filamentation⁶⁻⁸, white light generation⁹, terahertz production¹⁰, compression of ultrashort pulses¹¹, LiDAR¹², among others. In this work, we emphasized the nonlinear refractive index (n_2), due to its importance on the propagation of intense and ultrashort pulses, being its value necessary when considering the optical Kerr and self-phase modulation (SPM) effects.

Among the experimental techniques available employing a single wavelength for n_2 characterization in liquids and solids, Z-Scan¹³⁻¹⁴ and degenerate four-wave mixing (DFWM)¹⁵ are the commonly reported. However, they are quite troublesome for measuring any gaseous sample, because of the low density of the medium, resulting in a weaker NLO response than solid and liquid media (usually four orders of magnitude). Solely increase the peak power is not a proper solution for short interaction length, due to enhancement of plasma effects, which would masquerade a reliable n_2 value determination.

A technique based on SPM is the one often employed for n_2 characterization of gases¹⁶⁻¹⁷, which consists in the comparison of the pulse spectrum before and after the propagation through the medium. Nevertheless, there are others measurements employing optical Kerr effect¹⁸ and third-harmonic generation.¹⁹⁻²⁰ Others multi-beams techniques, such as pump-probe²¹⁻²³ and spectral interferometry²⁴⁻²⁵ require more complex experimental setups and analysis, though with the advantage of being suitable to study the material n_2 temporal dynamic.

Very recently, our group (Photonic's group – University of São Paulo) have developed an experimental setup for accurately measuring the nonlinear ellipse rotation (NER) effect and evaluating n_2 of liquid and solid materials.²⁶⁻²⁷ Additionally, we manage to measure this parameter as a function of the excitation pulse width (τ_p), from which we were able to evaluated and distinguish the amount of contribution coming from electronic nonresonant (ENR) and molecular orientation (MO) physical mechanism responsible for the material n_2 .²⁸ Furthermore, a comparison of NER and the well-established Z-Scan technique for the study of n_2 in solid and liquid were performed to demonstrate the reliability of the first. From that study,

we explore in deep relationship between the third order susceptibility ($\chi^{(3)}$) elements associated with each effect of those effects.²⁹

Motivated by the accomplishments obtained using NER to evaluate the solids and solvents n_2 , we conjectured the possibility to extend it for gaseous media confined in hollow core fiber (HCF), which opens the opportunity to study NLO properties of this important state of matter, so far weakly explored due the restrictions imposed by their low density.

An important application of gases n_2 characterization lays on the harmonic generation, for short wavelength generation, and spectrum broadening, for short pulse production. In those cases, ENR magnitude is an important asset of gases, which were explored during my internship at the Attosecond Ultrafast Optics group, at the Institute of Photonics Sciences (ICFO), Barcelona-Spain, for deep ultraviolet (DUV) generation.

Energetic and short source of laser light into a tunable DUV is for long desired, especially for their use in photolithography³⁰, spectroscopy³¹ and pump-probe measurements.³² This new source paves a way to a new regime of investigation. This goal motivates the usage of several NLO phenomena in order to produce such a source. One of the first efforts was to explore the four wave mixing (FWM) of a chirped-pulse amplified (CPA) Ti:Sapphire (790 nm, 1 kHz) laser as *Idler* and its second harmonic as a *Pump*.³³ By the time, it was generated 4 μ J at 1 kHz, in 270 nm, 10 fs of temporal width, characterized by self-diffraction (SD) frequency resolved optical gate (FROG)³⁴⁻³⁶, and applied into the temporally characterization of trans-, cis- internal conversion of a molecule.³⁷ Shortly after, a modified version of that experiment employing pump and idler chirped pulses (CFWM - chirped four-wave mixing) proved to produce higher pulse energies ($\sim 50 \mu$ J) and shorter temporal pulse width (sub-6 fs).³⁸⁻⁴⁰

Another effect used to produce short and energetic DUV was based on the spectral broadening, via SPM, of an amplified krypton fluoride (KrF) excimer laser (248 nm 110 fs e 20 mJ) in a HCF filled with a noble gas, followed by proper temporal compression^{11,41-46}. However, so far, the length of the HCF was the limiting factor, because it was only possible to produce a silica HCF as long as 2 m with accurate straightness of the inner hole. But, recently, this constrain has been overcome by the production of flexible HCF⁴⁷, which may be produced in a length up to several meters with proper conditions. Consequently, the longer HCF, the larger is the spectral broadening and shorter the Fourier-transformed limit (FTL) of the pulse. On this way, pulses with 200 μ J and 25 fs were generated in 248 nm⁴⁸, enhancing in one order of magnitude the DUV⁴⁹ pulse energy.

Besides of all progress made, there was still a necessity of an even higher energy source ($\sim 10 \mu$ J). In this sense, the effect of dispersive wave emission (DWE), observed on the dynamic

of high order optical solitons propagation in noble gas filled HCF has showed to be promising.⁵⁰ Under proper conditions, an intense, ultrashort, high order soliton may transfer energy into a blue shifted part of the spectrum via DWE, thus decaying into a lower order soliton. For a quite long time, SPM in HCF is employed to produce extremely short pulses (sub-10 fs) using mid IR amplified laser systems, though, within the parameters range employed by the time, the DWE phenomenon was never observed. Only after the energetic and ultrashort optical solitons were propagated in longer HCF, the DWE could be observed, and a reasonably amount of energy could be transferred to DUV. It was known the generated DUV preserved the temporal characteristics of its associated soliton, then DUV light with tunable central wavelength, controlled via gas pressure and/or coupled excitation pulse energy, and sub-10 fs of pulse width could be generated.

Nowadays, there is ongoing research on DUV generation via DWE using hollow core photonic crystal fibers (PCF).⁵¹⁻⁵⁴ The hollow core PCF have a very low loss due to its tiny diameter ($\approx 10 \mu m$), which is suitable for soliton propagation with an excellent control of NLO and dispersion properties, with the absence of undesired Raman or Rayleigh scattering effects.⁵⁵⁻⁵⁶ However, the HC-PCF cannot generate high energy DUV, because even considering its high transfer efficiency to DUV by DWE (10%), its manufacturing process limits its length into tens of cm, and due to its smaller inner diameter it supports much lower coupled pulse energy than nonstructured HCF, limiting the produced energy to hundreds of nJ. Evidently, those constraints made difficult the scaling of ultra-violet (UV) generation in HC-PCF, even though there is current efforts dedicated to improve it. For instance, the Kagome PCF architecture increase the UV pulse energy in one order of magnitude, compared to others architectures, where it has been demonstrated a tunable wavelength between 176 – 550 nm, with FTL bandwidth of tens of fs.⁵⁷⁻⁵⁸

Very recently, it was accomplished the production of DWE with very high energy (2 orders of magnitude above its predecessors) in long HCF (> 2 m), thus circumventing the limitations of the PCFs. A beam of 790 nm, 1 mJ e 10 fs was coupled into a Ne filled HCF of 220 μm of ID and 3 m of length, generating a tunable DUV between 130 – 350 nm and pulse energy around 5 to 8 μJ .⁵⁹ Optical gate and cross correlation techniques were used for the DWE temporal characterization, which revealed pulses width of 3 and 1.9 fs⁶⁰⁻⁶¹, respectively. Concomitant to our work, it was release a successful work on the generation of DUV, exploring the DWE in a great variety of parameters.⁶²

2 FIBER OPTICS

In this chapter, the basic theory of light propagation in optical fibers is presented. A brief introduction on step index fibers is given, exploring the basic phenomena and equations that are going to be useful when considering hollow core fibers, focusing on their propagation modes and dispersion. The main goals consist on the extension of the nonlinear light-gas interaction and the negative dispersion provided by the hollow core fibers, which are the essential topic to fully understand all experiments and results conducted in the thesis.

2.1 STEP INDEX FIBER

Conventional optical fibers have revolutionized the way light is transmitted through large distances. Due to the confinement, the light overcomes the diffraction of free space propagation, preserving its spatial mode at a very low loss.⁶³ The simplest architecture for this kind of light guidance is the step-index fiber (SIF), consisting of a transparent material in a cylindrical shape covered by a cladding made of the same material with a slightly lower refractive index. SIF architecture representation is depicted in Figure 1.

Figure 1 – Step index fiber profile.
Source: By the author.

In the framework of ray optics, the total internal reflection⁶⁴ is the physical mechanism responsible for the light propagation through the fiber. Its most important parameter is the normalized refractive index difference between the light propagating material core (n_c) and the external dielectric cladding (n_d) (Eq. (1)).

$$\Delta = \frac{n_c - n_d}{n_c}, \quad (1)$$

such as Δ must be positive for total internal reflection effect to take place.

The most common material employed in SIF manufacturing is fused silica (SiO_2), due to its suitable optical properties.⁶⁵ The refractive index difference is obtained introducing dopants on the fused silica matrix, which may increase its refractive index (core), or decrease it (cladding), according to the dopant material chosen.

Another parameter commonly characterized in optical fibers is the attenuation coefficient (α). The losses are modeled as the exponential decay of the impinging power of light (PW_0) while it propagates along the fiber ($PW(z)$, $0 \leq z \leq L$, where z is the propagation coordinate and L is the fiber length) (Eq. (2)).

$$PW(z) = PW_0 \exp(-\alpha z). \quad (2)$$

The losses in short wavelengths are due to inhomogeneities on refractive index of the fiber, on core-cladding interface, or concentration of impurities/defects which scatter light (Rayleigh, Raman and Brillouin scattering mainly) and so on. In longer wavelengths, the multiphoton absorption is the main loss effect.⁶⁶

2.1.1 Chromatic Dispersion

Chromatic dispersion is an essential concept when one considers light propagation through an optical fiber, and it manifests itself as a dependency of the refractive index on the light frequency (ω). By other means, dispersion implies that different spectral components of a light pulse do not travel through the medium at the same speed, and this has a huge impact on propagation, especially for spectral broadband laser pulses. The effect of chromatic dispersion is usually modeled by the Taylor development of pulse propagation wavenumber ($\beta(\omega) = n(\omega) \frac{\omega}{c}$) (3), n is the index of refraction and c is the vacuum speed of light) around the spectrum central frequency (ω_0). For the purposes of the phenomena reported on this work, only four terms were considered.

$$\beta(\omega) = \beta_0 + \beta_1(\omega - \omega_0) + \beta_2(\omega - \omega_0)^2 + \beta_3(\omega - \omega_0)^3 + \dots, \quad (3)$$

such as the zero order term (β_0) is constant and describes a common phase shift, which is independent of frequency; the first-order term (β_1) represents the reciprocal of group velocity (v_g) of the pulse envelope (Eq. (4)), and basically dictates how fast the pulse propagates through the material; while the second-order term (β_2) represents the chromatic group velocity dispersion (GVD) (Eq. (5)), then related with temporal pulse broadening, as the pulse frequency spreads temporally; the third-order term (β_3), also known as third-order dispersion (TOD), is of even weaker magnitude and is relevant only in a short range (few nanometers) around zero second order dispersion ($\beta_2 = 0$) but plays an important role when one considers to produce pulses.⁶⁷

$$\frac{d\beta}{d\omega} = \beta_1 = \frac{1}{v_g} = \frac{n_g}{c} = \frac{1}{c} \left(n + \omega \frac{dn}{d\omega} \right), \quad (4)$$

$$\frac{d^2\beta}{d\omega^2} = \beta_2 = \frac{1}{c} \left(2 \frac{dn}{d\omega} + \omega \frac{d^2n}{d\omega^2} \right). \quad (5)$$

The GVD plays a special role on light pulse propagation, and define two different propagation regimes, the *normal* and *anomalous* dispersion. The former, defined by the region where $\beta_2 > 0$, describes the regime where v_g decrease, while the optical frequency increase. In normal dispersion regime, the reddish region of the spectrum travels faster than the bluish one, and it is the case of the majority of transparent media in the visible range of spectrum. The latter defines the reciprocal effect ($\beta_2 < 0$ region) and it is of great importance to reverse the effects suffered by a spectral broadband optical pulse propagated by a material with normal dispersion. Generally, introduction of anomalous dispersion in an optical pulse spectrum range is tailored propagating it through a pair of prisms, gratings, or a set of dielectric dispersive mirrors, which is fundamental on the generation of ultrashort pulses.⁶⁸⁻⁶⁹

A pulse is said to be chirped whenever its instantaneous frequency of an optical pulse is time dependent. If $d\omega/dt > 0$ or < 0 , the pulse is said to be positively, or negatively chirped respectively. In particular, if the phase of the pulse is a quadratic function of time, the pulse is said to be linearly chirped. The practical meaning of the previous definition implies that the reddish part of a pulse spectrum travels in front of the blueish one for a positive chirp, or conversely for a negative chirp. The sign of the pulse chirp has a tremendous impact on the pulse propagation, because depending on the chirp sign and the dispersion regime, the optical pulse may be temporally broadened or compressed. Diffractive optics elements are manufactured, such as in each pulse interaction a certain amount of dispersion is introduced, in order to control the optical pulse temporal characteristics.

It is important to point out that the SIF core propagation wavenumber is slightly lower than the one of the core material, and slightly higher than the cladding material (Eq. (6)).

$$k_0 n_d < \beta < k_0 n_c, \quad (6)$$

such as $k_0 = 2\pi/\lambda_0$ is the vacuum propagation wavenumber.

Beyond the amount and kind of doping material, the geometry of the fiber may be chosen to control the value of frequency such as $\beta_2(\lambda_{zd}) = 0$ (λ_{zd} second order zero dispersion wavelength; 1.55 nm for the common telecommunication SIF), in comparison with the one of bulk material, thereby producing a dispersion suited to a specific application.

2.1.2 Propagation Modes

The light propagation must satisfy the Maxwell's equations (Eq. (7), (8), (9), and (10)).⁷⁰⁻⁷¹

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}, \quad (7)$$

$$\nabla \times \mathbf{H} = \mathbf{J} + \frac{\partial \mathbf{D}}{\partial t}, \quad (8)$$

$$\nabla \cdot \mathbf{D} = \rho_f, \quad (9)$$

$$\nabla \cdot \mathbf{B} = 0, \quad (10)$$

such as $\mathbf{E}$ and $\mathbf{H}$ are the electric and magnetic fields, respectively. $\mathbf{D}$ and $\mathbf{B}$ are the electric and magnetic flux densities, $\mathbf{J}$ and ρ_f are the current and charge density. As we are dealing with dielectric media we assume there is no free charges, then $\mathbf{J}$ and ρ_f are identical to zero, and the magnetic effects due to the optical field are too weak to be considered. Applying the free-charge assumption in the recurrence relations (Eq. (11-12)):

$$\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}, \quad (11)$$

$$\mathbf{B} = \mu_0 \mathbf{H} + \mathbf{M}, \quad (12)$$

such as ε_0 is the vacuum permittivity and μ_0 is the vacuum permeability. $\mathbf{P}$ and $\mathbf{M}$ are the induced electric and magnetic polarization density, respectively. For nonmagnetic media $\mathbf{M} = \mathbf{0}$.

After some algebra, we derive the electromagnetic wave equations that describe the light propagation through a medium (Eq. (13)).

$$\left[\nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right] \mathbf{E} = \mu_0 \frac{\partial^2 \mathbf{P}}{\partial t^2}, \quad (13)$$

such as $c = (\varepsilon_0 \mu_0)^{-1/2}$.

The left side of eq. (13) is the electromagnetic wave equation describes a propagation of light through vacuum, and the right side represent the source of the fields inside the medium. For a dielectric medium P may divided into two regimes, the linear ($\mathbf{P}^L$) and the nonlinear ($\mathbf{P}^{NL}$) ones.

$$\mathbf{P}(r, t) = \mathbf{P}^L(r, t) + \mathbf{P}^{NL}(r, t), \quad (14)$$

The two terms in the eq. (14) describe the linear and nonlinear optical regime. The light-matter interaction is said to be linear, whenever the induced polarization is linearly proportional to the optical electric field strength. For optical electric field magnitude close to the one that binds the outmost electron in the atom, or molecule, the nonlinear contribution must be

considered, up to the point the ionization phenomena start to dominate the optical phenomena. The nonlinear characteristics of the matter are developed in the chapter 3, and for the time being only the linear term in eq. (14) is going to be considered, introducing other basic characteristics of the SIF. The linear induced polarization (first term on eq. (14), $\mathbf{P}^L = \mathbf{P}^{(1)}$) is written as:

$$P_i^{(1)}(r, t) = \varepsilon_0 \int_{-\infty}^t \chi_{ij}^{(1)}(t - t') \cdot E_j(r, t') dt', \quad (15)$$

such as $\chi^{(1)}$ is a rank two tensor susceptibility. The eq. (15) is valid in the electric-dipole approximation and assume a local response of the medium.

It is often useful to represent the electromagnetic wave equation in the frequency domain applying the Fourier transformation ($\tilde{\mathbf{E}}(r, \omega) = \int_{-\infty}^{\infty} \mathbf{E}(r, t) \exp(i\omega t) dt$) to eq. (13).

$$\left[\nabla^2 + \varepsilon(\omega) \frac{\omega^2}{c^2} \right] \tilde{\mathbf{E}} = \mathbf{0}, \quad (16)$$

such as $\varepsilon(\omega) = (1 + \chi^{(1)}(\omega))\varepsilon_0$.

Expression (16) is known as the Helmholtz equation, and may be solved in cylindrical coordinates (usual symmetry for SIF manufacturing), resulting in:

$$\left[\frac{\partial^2}{\partial \rho^2} + \frac{1}{\rho} \frac{\partial}{\partial \rho} + \frac{1}{\rho^2} \frac{\partial^2}{\partial \phi^2} + \frac{\partial^2}{\partial z^2} + n^2(\omega) k_o^2 \right] \tilde{\mathbf{E}} = \mathbf{0}, \quad (17)$$

such as ρ and ϕ are the radial and polar coordinates, and z the longitudinal propagation coordinate.

As usual, E_z and H_z are chosen as the independent components, while the others come out from Maxwell relations. Equation (17) may be solved applying the separation of variables method (Eq. (18)).

$$\tilde{E}_z(r, \omega) = A(\omega) F(\rho) \exp(im\phi) \exp(i\beta z), \quad (18)$$

such as A and F are amplitudes depending on ω and ρ , respectively; m is an integer number. For the sake of simplicity, it has been assumed no change in the optical electric field throughout propagation; hence A is independent of z -coordinate. $F(\rho)$ is a solution of the Bessel equation.

$$\left[\frac{d^2}{d\rho^2} + \frac{1}{\rho} \frac{d}{d\rho} + n^2(\omega) k_o^2 - \beta^2 - \frac{m^2}{\rho^2} \right] F(\rho) = 0, \quad (19)$$

whose solution are the well-known Bessel functions, being the first kind solution (J_m) the one with physical meaning, and the second order (K_m) given for completeness. If the losses are small and could be neglected, we may use $\varepsilon(\omega) = n^2(\omega)$ in eq. (19). Thus, the solution for the propagation distribution inside the core (Eq. 20) and cladding (Eq. 21) region are:

$$F(\rho) = J_m(\kappa\rho), \text{ for } \rho \leq a; \quad (20)$$

$$F(\rho) = K_m(\sigma\rho), \text{ for } \rho > a; \quad (21)$$

such as:

$$\kappa = (n_c^2 k_0^2 - \beta^2)^{\frac{1}{2}}, \quad (22)$$

$$\sigma = [-(n_d^2 k_0^2 - \beta^2)]^{1/2}, \quad (23)$$

such as κ and σ being the transverse propagation wavenumber of the core (Eq. (22)) and cladding (Eq. (23)), respectively.

Equation (20) represent the spatial distribution modes that are able to propagate along the core region. Likewise, eq.(21) is the field distribution along the cladding region, which do not guide the pulse of light and its solution models an exponential decay.

Figure 2 – SIF propagation wavenumber.
Source: By the author.

Across the core-cladding interface ($\rho = a$; where a is the core radius) the field components in the radial and polar coordinates must be continuous, leading to the follow eigenvalue equation.

$$\left[\frac{J'_m(\kappa a)}{\kappa J_m(\kappa a)} + \frac{K'_m(\sigma a)}{\sigma K_m(\sigma a)} \right] \left[\frac{J'_m(\kappa a)}{\kappa J_m(\kappa a)} + \frac{n_d^2}{n_c^2} \frac{K'_m(\sigma a)}{\sigma K_m(\sigma a)} \right] = \left(\frac{m\beta k_o(n_c^2 - n_d^2)}{an_c \kappa^2 \sigma^2} \right), \quad (24)$$

such as the prime is the derivative with respect to the argument. Additionally, the eq. (25) was employed.

$$\kappa^2 + \sigma^2 = k_o(n_c^2 - n_d^2). \quad (25)$$

The eigenvalues for β (eq. (24)) give the possible values to the propagation wavenumber for each value of m . In order to include the several zeros of the Bessel function ($J_m(u_{mn}) = 0$, where u_{mn} is the n -th root of the m -th order), another index is added to the propagation wavenumber (β_{mn}). Both indexes, m and n , represent each individual mode supported by the SIF. Consequently, the field distribution is obtained by eq. (18). The modes propagated in the optical fiber are generally hybrid electric (EH_{mn}) or magnetic (HE_{mn}), but when $m = 0$ they

fall into the transverse-electric (TE_{mn}) and –magnetic (TM_{mn}) mode of a planar waveguide, for the azimuthal componets of the fields vanishes. The number of modes supported by the fiber (N_m), at a given wavelength, depend on its design geometric parameters (i.e. radius for cylindrical geometry) and the refractive index difference. A useful parameter, called V -number, is often used to define the amount of modes allowed to propagate in a SIF, and it is a radius normalized frequency when $\sigma = 0$.

$$V = \kappa a = k_0 a \sqrt{n_c^2 - n_d^2}, \quad (26)$$

such as κ is given by eq. (25). Concisely, N_m relates to V by:

$$N_m \approx \frac{1}{2} V^2 \quad (27)$$

Equation (27) shows that the larger the radius or the refractive index difference, the bigger is amount of modes supported by SIF fiber. Conversely, there is a lower limit, down to the point where only one mode is allowed to propagate. All the other modes, but the fundamental, are forbidden if $V < V_c$, such as $J_0(V_c) = 0$ or $V_c = u_{00} \approx 2.405$. Accordingly, the optical fibers that satisfy such condition are said single mode fibers.

2.1.3 Interaction Length Extension

Another important characteristic that makes propagation through fibers fundamental to the phenomena studied in this work is the extension of the Rayleigh range (z_R) of a laser beam. Light propagation through fibers allow a considerably increasing of interaction length with the core-filled material in a high intensity regime, particularly useful for NLO optics studies and applications. The Rayleigh length of a focused light beam, by definition, is the distance in which the beam radius at focus (w_0) is increased by a factor of $\sqrt{2}$ along the propagation direction. For light beams with gaussian spatial profile, the Rayleigh length is given by:

$$z_R = \frac{\pi w_0^2}{\lambda}, \quad (28)$$

such as $\lambda = \lambda_0/n$.

Essentially, the confocal parameter ($2z_R$) determines the depth of a focused beam, where the wave front may be considered plane and the light intensity (I , light power per area) may be assumed constant. For nonlinear optics purposes, the light intensity must be such as its corresponding electric field is at the same magnitude order than the one that binds valence electrons into atoms or molecules. The previous condition is usually accomplished by focusing a pulsed light beam, because the smaller w_0 becomes, the higher is the light intensity. By this

way, nonlinear process in bulk media are proportional to $I_0 z_R$, where there is a tradeoff between w_0 and z_R for focused beams, which limits one or the other. Albeit, this limitation can be overcome by propagating the beam through a fiber, then increasing z_R while preserving w_0 .

2.2 HOLLOW CORE FIBERS

Hollow core fibers, also known as capillaries, are fibers where the core material is absent, being the cavity free to be filled with nonsolid materials (gases, liquids, etc.), or evacuated. In HCF, the core index of refraction is may be lower than the cladding material (usually silica). Consequently, HCFs light transmission is no longer based on total internal reflection effect, but, instead, in a grazing incidence of coupled light. Their main advantages consist on the fact that its damage threshold is higher than step index fibers, and their broadband spectrum transmission capabilities. In addition, the HCF geometry or filling material may be chosen such as to tailor the system dispersion properties.

2.2.1 Dispersion Control

Control the light dispersion characteristics in HCF is possible through the combination of the modal dispersion imposed by the waveguide geometry and the one of the core-filling material. The often-adopted model for a HCF optical dispersion is the Marcatili-Schmeltzer (MS).⁷²⁻⁷⁴ The MS model assumes an effective mode propagation wavenumber (β_{eff}) associated to an effective refractive index ($n_{2,eff}$).

$$\beta_{eff} = k_0 n_{eff}, \quad (29)$$

Substituting eq. (29) into eq. (22), we obtain the transverse propagation wavenumber of HCF. Assuming the entrance propagation wavenumber comes from a gaseous atmosphere, we obtain:

$$k_0 n_{gas}^2 = \beta_{eff}^2 + \kappa^2, \quad (30)$$

substituting eq. (29) into eq. (30) we have the transversal propagation wavenumber:

$$\kappa = k_0 \sqrt{n_{gas}^2 - n_{eff}^2}. \quad (31)$$

such as n_{gas} is the gas refractive index.

The eq. (24) may be simplified whenever the following conditions are true:

$$k_0 a \gg n_d u_{mn}, \quad (32)$$

$$\beta/k - 1 \ll 1, \quad (33)$$

such as the first condition states the HCF radius is much larger than the free space wavelength, whereas the second inequality restrict the analysis to low-loss modes, in the sense the β is nearly k_0 , which is a reasonable whenever the HCF is filled with gases. Under those constraints, and using the asymptotic properties of the Bessel functions, the eigenvalue equation (eq. (24)) may be rewritten as:

$$J_m(\kappa a) = i \left[\frac{n_d^2 + 1}{2\sqrt{n_d^2 - 1}} \right] \frac{\kappa}{k_0} J_{m+1}(\kappa a), \quad (34)$$

For the sake of simplicity, it is adopted the perfect conductor approximation, which considers the transverse guided core modes are zero at core-cladding interface. In other word, the previous condition states that $J(\kappa a) = 0$, so $\kappa a = u_{mn}$. Substitution κa argument in eq. (31), the capillary model then predicts that the effective mode index of the HCF is:

$$n_{eff}^2 = n_{gas}^2 - \frac{u_{mn}^2}{a^2 k_0^2}. \quad (35)$$

Linear absorption in optical frequencies may be neglected in gaseous media, and only the real part of its linear susceptibility need to be accounted (Eq. (36)).

$$n_{gas}^2 = 1 + \chi_{gas}^{(1)}(\omega). \quad (36)$$

Furthermore, the linear susceptibility of gases scales with the density of gaseous particles/molecules (N), and for ideal gases $N/N_0 = pT_0/(p_0T)$, such that the p_0 and T_0 are the pressure and temperature in the standard condition ($T_0 = 273$ K and $p_0 = 1$ atm). Consequently, eq. (36) is rewritten as:

$$n_{gas}^2(\omega, p, T) = 1 + \frac{p}{p_0} \frac{T_0}{T} \chi_{gas}^{(1)}(\omega) \Big|_{p_0, T_0}. \quad (37)$$

The capillary model to the propagation effective wavenumber for a HCF is obtained substituting eq. (36) into eq. (35), and its result eq. (29).

$$\beta_{eff}(p, T, \omega) = k_0 \sqrt{1 + \frac{p}{p_0} \frac{T_0}{T} \chi_{gas}^{(1)}(\omega) \Big|_{p_0, T_0}} - \frac{u_{mn}^2}{a^2 k_0^2}. \quad (38)$$

The right hand side of eq. (38) is developed in Taylor series around one, in order to separate the gas and capillary contribution, then it is straightforward to obtain the relations for HCF propagation phase velocity (ω_0/β_0), group velocity ($1/\beta_1$), GVD (β_2) and the third order dispersion (β_3). For the purposes of this work, the GVD is the one worth to discuss. From Eq. (38):

$$\beta_{2,mn}(p, T, \omega) \approx \frac{1}{c} \frac{p}{p_0} \frac{T_0}{T} \left(\frac{\partial}{\partial \omega} + \frac{\omega}{2} \frac{\partial^2}{\partial \omega^2} \right) \chi^{(1)}(\omega) - \frac{u_{mn}^2 c}{a^2 \omega^3}. \quad (39)$$

The left term on the right hand side of eq. (39) depicts the dispersion of the HCF filling gas, which is linearly proportional with the pressure and inversely with the gas temperature, whereas the right term represent the waveguide negative contribution to dispersion. It depends inversely on the square radius of the capillary, which is an exclusive characteristic of the waveguide geometry. Even though it may seem suited to work with small radius, in order to enhance the negative HCF dispersion, their losses and damage threshold also become higher. Then, HCF internal diameter larger than 100 μm are used to couple large amounts of light energy, leading to very interesting applications as high harmonic generation (HHG)², generation of fs-pulses in the deep ultraviolet (DUV)⁶², and soliton self-compression.⁷⁵ Notably, an empty HCF presents a negative dispersion, and filling the HCF cavity with a noble gas, in a controllable manner, we are able to tune λ_{zd} . For the conditions presented in this work, λ_{zd} can be tuned from ultraviolet to near infrared.

2.2.2 Losses

A theoretical expression for HCF losses could be obtained neglecting the perfect conductor approximation.⁷² Although laborious, it uses the low-loss condition (Eq. (33)), which makes the right hand side of eq. (34) be close to zero, then using a perturbation technique and keeping only the first term, results in:

$$\kappa a \approx u_{mn} \left[1 - \frac{i}{k_0 a} \left(\frac{n_d^2 + 1}{2\sqrt{n_d^2 - 1}} \right) \right]. \quad (40)$$

Substituting this relation on eq.(22) does not change the real part of the propagation wavenumber (Eq. (38)), but the imaginary part of it gives the HCF attenuation constant.

$$\alpha_{mn} = \left(\frac{u_{mn}}{2\pi k_0} \right)^2 \frac{1}{a^3} \left(\frac{n_d^2 + 1}{2\sqrt{n_d^2 - 1}} \right). \quad (41)$$

Attenuation in HCF is related to the reciprocal of $k_0^2 a^3$, meaning the larger the radius, the smaller the losses. Therefore, there is a tradeoff of how big the radius should be, because it can enhance the transmission efficient in detrimental to the magnitude of the negative dispersion that could be introduced. Then, the suitable radius should be chosen accordingly to the desired properties and application.

3 NONLINEAR OPTICS

Chapter 3 introduces all the theory on the nonlinear optics phenomena necessary in this work. It starts giving the basic concept of nonlinear polarization, which pave the way of all nonlinear phenomena, with emphasis on the third order susceptibility in isotopic centrosymmetric media, which is the case of gases. Additionally, we present the derivation of the nonlinear refractive index, one of the major subjects of this work. The chapter evolves exploring the nonlinear circular birefringence, which is the physical basis behind the nonlinear ellipse rotation effect, and establishing the fundamental relation among the nonlinear third-order tensor elements and n_2 . Following, the nonlinear Schrödinger equation is derived for explaining the nonlinear light propagation through a hollow core fiber, and the phenomena of self-phase modulation and optical soliton dynamics.

3.1 NONLINEAR POLARIZATION

The optical response of a media becomes nonlinear whenever the induced polarization on atoms and molecules in the matter no longer scales linearly with the optic electric field. Up to a certain point, NLO effects are treated as a weak perturbation on the harmonic motion of bound valence electrons induced by optical frequencies, i.e. the anarmonic oscillations. As a rough estimation, NLO phenomena start to manifest their effects when optical electric field is comparable with the one that binds the outmost electron in the atoms that constitute the media (e.g. $\approx 5 \times 10^{11} \text{V/m}$ for hydrogen atom). As any perturbation, deviations from nonperturbed values could be obtained through a power series development, thereby $\mathbf{P}^{NL}$ in eq. (14) can be written as:

$$\mathbf{P}^{NL} = \mathbf{P}^{(2)} + \mathbf{P}^{(3)} + \dots, \quad (42)$$

such as $\mathbf{P}^{(2)}$ e $\mathbf{P}^{(3)}$ are the second and third order induced polarizarion. In terms of their components, the electronic nonlinearities are described by:

$$P_i^{(2)}(r, t) = \varepsilon_0 \iint_{-\infty}^t \chi_{ijk}^{(2)}(r, t - t_1, t - t_2) : E_j(r, t_1) E_k(r, t_2) dt_1 dt_2 \quad (43)$$

$$P_i^{(3)}(r, t) = \varepsilon_0 \iiint_{-\infty}^t \chi_{ijkl}^{(3)}(r, t - t_1, t - t_2, t - t_3) : E_j(r, t_1) E_k(r, t_2) E_l(r, t_3) dt_1 dt_2 dt_3 \quad (44)$$

$\chi^{(n)}$ is a $n + 1$ rank electric susceptibility tensor with 3^{n+1} elements that depends on spatial coordinates and optical frequency, and the sub-indexes $i, j, k, l = x, y, z$ corresponding any of the spatial coordinates. Each following term on the electrical susceptibility power series

development (Eq. (42)) is smaller than the previous term by several orders of magnitude (e.g. $\chi^{(2)}/\chi^{(3)} \approx 10^{12} \text{ V}^2/\text{m}^2$). Consequently, the second order induced polarization only becomes relevant whenever $\chi^{(2)}E^2$ ($I \propto E^2$) is comparable with $\chi^{(1)}E$, and so on. The advent of nonlinear optics came soon after the development of the laser, because only then it was possible to generate light intensities high enough to observe NLO phenomena.

Several physical mechanisms induce to optical nonlinearities. They may come out from a pure displacement of the atomic electron cloud (Eq. (43) and (44)) happening at the femtosecond time scale; atomic nuclear motion at picoseconds; charge distribution throughout a conjugated molecule, from population effects on molecules energy levels at nanoseconds, heat transfer at milliseconds, or manifestation of plasma effects. Whenever the response of the NLO phenomena is two orders of magnitude faster in comparison with the duration of the excitation, the response could be considered instantaneous from the time point of view. In the case of instantaneous effects, eq. (43) and (44) suffer a simplification by adding a Dirac's delta functions ($\delta(t - t_i)$) for each electric field component.

Higher-order NLO effects (beyond $\chi^{(3)}$) are rather seldom reported, because the necessary light intensity at which $\mathbf{P}^{(4)}$ would get close to $\mathbf{P}^{(1)}$ is commonly above the atom or molecule ionization threshold. Additionally, as far as we know, there is no how to suppress $\mathbf{P}^{(3)}$ effects, which becomes is dominant over any other in isotropic and centro-symmetric media.⁷⁶⁻

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3.1.1 NLO Susceptibility in Isotropic Media

Every single term in the expansion of the NLO polarization (Eq. (42)) is responsible for several NLO optical effects. For instance, $\chi^{(2)}$ is linked to sum (SFG) and difference (DFG) of frequency generation, optical parametric oscillation (OPO), among others. On the other hand, $\chi^{(3)}$ is the one related with third-harmonic generation (THG), intensity-dependent refractive index and self-focusing, NLO Raman, SPM, and FWM. However, the symmetries of a NLO medium impose restriction on the effects that may or not happen. For the case of noble and simple gases employed in this work, the medium is homogenous and isotropic, thus symmetry considerations simplify even further the expression of $\mathbf{P}^{NL}$.⁷⁹

Any medium symmetry dictates its physical properties must remain invariant under their respective transformation operation, as in the case of centrosymmetric media, which possess inversion symmetry (S_i). In practice, S_i change the sign of all Cartesian components, which results in its transformation rotation matrix (R_{ij}) being identical to a minus Kronecker's delta

$(-\delta_{ijk})$. Consequently, applying the inversion symmetry operation at $\chi^{(2)}$, which transform itself as a third-rank tensor, we have:

$$\chi'_{ijk} = S_i \chi_{\alpha\beta\gamma} = (-\delta_{i\alpha})(-\delta_{j\beta})(-\delta_{k\gamma})\chi_{\alpha\beta\gamma} = -\chi_{ijk}, \quad (45)$$

which the obvious solution is $\chi_{ijk}^{(2)} = 0$, and then $\mathbf{P}^{(2)} = \mathbf{0}$. The same is valid to all others even order members of the induced polarization ($\mathbf{P}^{(4)}$, $\mathbf{P}^{(6)}$, ...) of the electrical susceptibility development. Thus, $\chi_{ijkl}^{(3)}$ is the first NLO term that prevails.

As it is the case of atomic gases, isotropic media have mirror symmetry on the planes xy (σ_{xy}), xz (σ_{xz}), and yz (σ_{yz}); 90° rotation symmetry around the axis x ($C_{90^\circ}^x$), y ($C_{90^\circ}^y$), z ($C_{90^\circ}^z$) and 45° around z -axis (C_{45°). As it happens, symmetry considerations may turn the $\chi^{(3)}$ components null or identical among themselves. At first glance, $\chi_{ijkl}^{(3)}$ is a fourth-rank tensor possessing 81 elements, but when mirror symmetries are applied on $\chi^{(3)}$, they prevent that any component, whose the index (i, j, k, l) appear an odd number of times, be different from zero. This last statement reduces to 21 the number of components in $\chi^{(3)}$. Moreover, when 90° rotation symmetry is applied, several components become identical, ending up on the following identities:

$$\begin{aligned} \chi_{xxyy} &= \chi_{zzxx} = \chi_{yyzz} = \chi_{zzyy} = \chi_{yyxx} = \chi_{xxzz}; \\ \chi_{xzzx} &= \chi_{zxzx} = \chi_{yzyz} = \chi_{zyyz} = \chi_{yxxxy} = \chi_{xyyx}; \\ \chi_{xyxy} &= \chi_{zxzx} = \chi_{yzyz} = \chi_{zyyz} = \chi_{yxyx} = \chi_{xzzx}; \\ \chi_{xxxx} &= \chi_{yyyy} = \chi_{zzzz} = \chi^*; \end{aligned} \quad (46)$$

which are related by the 45° z -axis rotation symmetry:

$$\chi_{ijkl} = \chi_{xxyy}\delta_{ij}\delta_{kl} + \chi_{zyyz}\delta_{il}\delta_{jk} + \chi_{xyxy}\delta_{ik}\delta_{jl}, \quad (47)$$

Then, from the previous 21 elements, three showed up as linear independent after mirror and rotation operation.⁷⁷

In addition another symmetry applies, it is know as full intrinsic permutation, that does not have physical meaning because it is based on a mathematical argument valid ever since the medium is lossless, allows any χ_{ijkl} index to be permuted once its correspondent frequency term is permuted on the same way (e.g. $\chi_{ijkl}(\omega_\alpha; \omega_\beta, \omega_\gamma, \omega_\nu) = \chi_{ijlk}(\omega_\alpha; \omega_\beta, \omega_\nu, \omega_\gamma)$). Then, for effect induced by a single optical beam, where the frequencies of all photons are the same, it decreases even more the degree of freedom of $\chi^{(3)}$ components, resulting in the equality of the terms χ_{xxyy} and χ_{xyxy} . It is conventional to call the remaining two linearly independent components as $A = 6\chi_{xxyy}$ and $B = 6\chi_{xyyx}$, which are linked to the isotropic and anisotropic

components of $\chi^{(3)}$ tensor.⁸⁰⁻⁸² In general, A and B have the same sign (positive), but some liquid samples may present opposite signs.⁸²

Finally, attending all the conditions, eq. (47) is rewritten as:

$$\chi^* = \frac{1}{3} \left(A + \frac{B}{2} \right). \quad (48)$$

In order to give a more simplified picture of the $\chi^{(3)}$ tensor, after all symmetry properties for centrosymmetric and isotropic media described, an index contraction is employed, resulting in a bidimensional matrix depicted in eq. (49).

$$\chi^{(3)} = \begin{pmatrix} & xx & yy & zz & yz & xz & xy \\ xx & \chi^* & A & A & 0 & 0 & 0 \\ yy & A & \chi^* & A & 0 & 0 & 0 \\ zz & A & A & \chi^* & 0 & 0 & 0 \\ yz & 0 & 0 & 0 & B & 0 & 0 \\ zx & 0 & 0 & 0 & 0 & B & 0 \\ xy & 0 & 0 & 0 & 0 & 0 & B \end{pmatrix}. \quad (49)$$

Beyond the symmetries considerations, that reduce all the 81 components of $\chi_{ijkl}^{(3)}$ into two linear independent ones (A and B), there is a proportionality between A and B which depends on the physical mechanism that induces $\mathbf{P}^{(3)}$, that originates from empirical result corroborated by several experiments⁸² and followed by theoretical models.⁷⁶ The B/A is 1 when $\mathbf{P}^{(3)}$ is induced by electronic nonresonant (ENR), and 6 when $\mathbf{P}^{(3)}$ is induced by molecular orientation (MO) effect.⁸³

3.1.2 Intensity Dependent Refractive Index and Absorption

One of the remarkable effects of $\chi^{(3)}$ is the light induced change of the medium refractive index⁸⁴, also known as optical Kerr effect. Such change is characterized by the nonlinear refractive index (n_2), which represent the coefficient of the refractive index linear variation ($\delta n = n(I) - n_0$) with the light intensity (Eq. (50)), which only occur at the NLO regime. In addition, the imaginary part of $\chi^{(3)}$ give rise to an intensity dependent absorption coefficient ($\delta \alpha = \alpha(I) - \alpha_0$) in a lossy media (Eq. (51)).

$$\delta n = n_2 I, \quad (50)$$

$$\delta \alpha = \alpha_2 I, \quad (51)$$

hence forward, we adopt the light intensity definition as: $I = 2n_0 \varepsilon_0 c |\mathbf{E}(\omega)|^2$.

In order to associate the change in the refractive index and absorption (measurable quantities) with $\chi^{(3)}$, we rewrite eq. (14) in the frequency domain, for a single and linearly polarized beam⁷⁶:

$$\mathbf{P}(\omega) = \varepsilon_0 \chi^{(1)} \mathbf{E}(\omega) + 3\varepsilon_0 \chi^*(\omega) |\mathbf{E}(\omega)|^2 \mathbf{E}(\omega), \quad (52)$$

For convenience, we define an effective susceptibility and refractive index from eq. (52):

$$\chi_{eff} = \chi^{(1)} + 3\chi^{(3)} |\mathbf{E}(\omega)|^2, \quad (53)$$

$$n_{eff}^2 = 1 + \chi_{eff}. \quad (54)$$

By substitution of eq. (50) and (53) in eq. (54), and truncating to terms of quadratic order ($|\mathbf{E}(\omega)|^2$), results in:

$$n_2 = \frac{3}{4n_0^2 \varepsilon_0 c} \text{Re}(\chi^{(3)}), \quad (55)$$

The eq. (55) links the nonlinear refractive index with the real part of the third order susceptibility, and by a similar analysis, we reach an expression for the NLO absorption coefficient (Eq. (56)).

$$\alpha_2 = \frac{3\omega_0}{2n_0^2 \varepsilon_0 c^2} \text{Im}(\chi^*). \quad (56)$$

NLO refractive index and absorption coefficient are very important parameters, especially when one considers the propagation of an intense light pulse through a medium. The literature is full of measurements of n_2 , performed with several kinds of techniques, wavelength and pulse duration for several materials.⁸⁴⁻⁸⁶

3.2 NONLINEAR BIREFRINGENCE

A major simplification is done whenever light preserves its polarization state along the fiber propagation. Even though we are working with isotropic media, in which there is no permanent birefringence, and considering there is no stress and core-shape variations, there may be a manifestation of an intensity dependent coupling between two orthogonally degenerate polarized modes, in the NLO regime, also known as cross-phase modulation (XPM). Here, we treated the case where the NLO birefringence is induced by a single beam, where all photons involved possess the same central frequency.⁸⁷

3.2.1 Cartesian Basis

In the linear basis, the third-order-induced polarization for a self-induced effect can be written as:

$$\mathbf{P}^{(3)}(\omega; \omega, \omega, -\omega) = \frac{1}{2} \left(P_x^{(3)} \hat{\mathbf{x}} + P_y^{(3)} \hat{\mathbf{y}} \right) \exp(-i\omega_0 t). \quad (57)$$

Rewriting $\mathbf{P}^{(3)}$ in eq. (57), in order to put each cartesian component in evidence, we have⁷⁶⁻⁷⁷:

$$P_i^{(3)}(\omega) = 3\varepsilon_0\chi_{ijkl}(\omega; \omega, \omega, -\omega)E_j(\omega)E_k(\omega)E_l(-\omega). \quad (58)$$

Then, substituting the eq. (47) in (58) and considering solely an ENR effect ($A = B$ or $\chi_{xxyy}^{(3)} = \chi_{xyxy}^{(3)} = \chi_{xyyx}^{(3)}$), we obtain the following relation:

$$P_i^{(3)} = 3\varepsilon_0\chi^* \left[\left(|E_i|^2 + \frac{2}{3}|E_j|^2 \right) E_i + \frac{1}{3}(E_i^* E_j) E_j \right]. \quad (59)$$

The last term in eq. (59) is related with a FWM process, and it is not going to be considered. However, the first term is proportional to E_i and, by the same procedure employed in eq. (55), it is associated with a value of n_2 . Then, we have:

$$\delta n_i = 2n_0\varepsilon_0cn_2 \left(|E_i|^2 + \frac{2}{3}|E_j|^2 \right). \quad (60)$$

The eq. (60) represent the nonlinear refractive index variation in each of the propagation transverse coordinates, where its first term on the right side is responsible for SPM, self-focusing, and others n_2 related effects. However, the second term characterizes the XPM, and represents the NLO phase shift acquired by one polarization component induced by other. As δn_x and δn_y may be unequal, depending on the polarization state, it creates an NLO birefringence on the medium, whose magnitude depends on the optical intensity. In practice, XPM manifest itself as a rotation of an elliptical polarized beam (Figure 3), and the phenomenon is referred to as nonlinear ellipse rotation (NER).⁸⁸

Figure 3 – Representation of the NER effect, depicting the change suffered by an (a) incident elliptical polarization state and after it has been (b) transmitted through a $\chi^{(3)}$ medium.

Source: Adapted from BOYD.⁷⁶

3.2.2 Circular Basis

An alternative, and more intuitive, way to represent the NER effect consist of rewrite the NLO induced polarization and the optical electric field in terms of a circular basis. Analogous to the Cartesian base, the polarization state can be described as a linear combination of circular basis elements:

$$\mathbf{P}^{(3)} = P_+^{(3)}\hat{\mathbf{e}}_+ + P_-^{(3)}\hat{\mathbf{e}}_-, \quad (61)$$

in the same way:

$$\mathbf{E} = E_+\hat{\mathbf{e}}_+ + E_-\hat{\mathbf{e}}_-, \quad (62)$$

such as $\hat{\mathbf{e}}_{\pm} = (\hat{\mathbf{x}} \pm i\hat{\mathbf{y}})/\sqrt{2}$ correspond to a counterclockwise (+) and clockwise (-) unitary vectors, where the rotatory direction is defined as seen by an observer looking towards counter propagation direction.

In order to generalize $\mathbf{P}^{(3)}$ for an isotropic media, it is substituted eq. (47) into eq. (58), which results in:

$$\mathbf{P}^{(3)}(\omega; \omega, \omega, -\omega) = \varepsilon_0 A(\mathbf{E} \cdot \mathbf{E}^*)\mathbf{E} + \frac{1}{2}\varepsilon_0 B(\mathbf{E} \cdot \mathbf{E})\mathbf{E}^*. \quad (63)$$

Now, substituting eq. (61) and (62) into eq. (63) we reach in a convenient third order susceptibility expression for each right- and left-handed polarization state ($P_{\pm} = \epsilon_0 \chi_{\pm}^{(3)} E_{\pm}$) in terms of A and B :

$$\chi_{\pm}^{(3)} = A|E_{\pm}|^2 + (A + B)|E_{\mp}|^2, \quad (64)$$

such as this circular third-order susceptibility already includes a squared optical electric field.

Associating this $\chi_{\pm}^{(3)}$ to an electric permissivity ($\varepsilon_{\pm} = \varepsilon_0(1 + \chi^{(1)} + \chi_{\pm}^{(3)})$) and refractive index ($n_{\pm} = \sqrt{\varepsilon_{\pm}}$) on the same circular basis. We have:

$$n_{\pm} = n_0 \sqrt{1 + \chi^{(1)} + \chi_{\pm}^{(3)}} \approx n_0 + \frac{1}{2n_0} [A|E_{\pm}|^2 + (A + B)|E_{\mp}|^2], \quad (65)$$

such as the power series was done under the criteria $\chi_{\pm}^{(3)} \ll 1 + \chi^{(1)}$.

By this way, the medium induced circular birefringence is defined as:

$$\Delta n = n_+ - n_- = \frac{B}{2n_0} (|E_-|^2 - |E_+|^2), \quad (66)$$

noticing Δn depends only on B .

It is useful to express $|E_{\pm}|$ in terms of Cartesian basis that is easier assessed⁸⁹:

$$|E_{\pm}|^2 = \frac{(1 \pm \tan(\varphi))^2}{2(1 + \tan^2(\varphi))} |E|^2, \quad (67)$$

such as the eccentricity (φ) of the elliptical polarization state is defined as:

$$\tan^2(\varphi) = \frac{|E_{min}|^2}{|E_{max}|^2}, \quad (68)$$

and it is the parameter that associated to the light polarization state, varying between zero (linearly polarized state) and $\pi/4$ (circularly polarized state).⁹⁰⁻⁹¹

The induced circular birefringence is associate to a cross phase-shift ($\Delta\Phi = k_o\Delta nL$) between the two circular components, and after substituting eq. (67) and (68) into (66), result in eq. (69):

$$\Delta\Phi = k_o \left[\frac{B \sin(2\varphi)}{4n_0^2 \epsilon_0 c} \right] LI. \quad (69)$$

3.3 NONLINEAR ELLIPSE ROTATION

The effect of NER, as explained in the section 3.2, manifest itself as rotation of the symmetry axis that define an elliptically polarized beam, without any change on its eccentricity (Figure 3), when it propagate through a Kerr medium in the NLO regime.

It is important to make distinction of the NER effect from the Faraday effect and the optical activity, because each one of them have a completely different physical origins, though all of them rotate the initial polarization state of light.^{64,76} The Faraday effect require a much more intense magnetic field than the one provide by light, even in the NLO regime. The optical activity manifest itself also as a circular birefringence, though it is a $\chi^{(1)}$ effect of quiral molecules and does not depend of the electric field amplitude. Essentially, NER is a self-induced birefringence coupling of two orthogonal components that define the polarization state. Although the Kerr effect is also another example of self-induced birefringence, there is no coupling between the orthogonal fields, which distinguish it from NER effect.

The model describing NER attributes its effects to the NLO third-order susceptibility ($\chi^{(3)}$)⁸¹, in such a way that its tensorial nature must be taken into account. Distinct physical mechanism induce $\chi^{(3)}$ effects. Pure $\chi^{(3)}$ effects are only due to small harmonic displacement of the atomic or molecular electronic cloud, though the theory is expanded to accommodate other effects that generate similar optical responses, as the nuclear contribution. The mechanisms that contribute to NLO induced birefringence into the medium are of electronic nonresonant, resonance signal enhancement close to a Raman or two-photon absorption transition, electrostriction, molecular orientation or redistribution, thermal variation, or Soret

effect origins.^{76,77,90} The electronic nonresonant and molecular orientation are the most relevant to the NER effect on the parameters range of this study.

3.3.1 Polarization Dependency

The NER is a phenomenon that manifest itself only when the polarization state of the optical field is elliptical and depend only on the B coefficient of $\chi^{(3)}$. From eq. (66) it can be realized neither a propagating linearly ($|E_-| = |E_+|$) nor circular ($|E_-|$ or $|E_+|$ is equal to zero) polarized optical field induces circular birefringence (i.e. $\Delta n = 0$). Nonetheless, it is still possible to define an index of refraction variation ($\delta n = n_{\pm} - n_0$, eq. (65)). For the linear case ($\tan(\varphi) = 0$), both components contribute equally to the cross phase shift, then:

$$\delta n_{linear} = \frac{1}{2n_0} \left(A + \frac{B}{2} \right) |E|^2, \quad (70)$$

such as $|E|^2 = 2|E_{\pm}|^2$ is the total optical field. For the circularly polarization state ($\tan(\varphi) = 1$), then:

$$\delta n_{circular} = \frac{1}{2n_0} A |E|^2. \quad (71)$$

The equations (70) and (71) demonstrate that whenever A and B have the same sign, the index of refraction variation is greater for a linear than for a circular polarized state. Furthermore, the evaluation of the ratio between the index of refraction variation on those polarization states is useful to determine which physical mechanism is predominant on the medium⁹², because:

$$\frac{\delta n_{linear}}{\delta n_{circular}} = 1 + \frac{B}{2A} = \begin{cases} 3/2 \rightarrow ENR \\ 4 \rightarrow OM \end{cases}, \quad (72)$$

such as the B and A ratio for pure ENR, or with MO contribution was called.

3.3.2 Accessing B

As demonstrated on section 3.2.2, the circular birefringence only occur when the optical field is in an elliptical polarization state, and the difference between the refractive index for each orthogonal basis is direct related to the B component of $\chi^{(3)}$. In turn, the B parameter can be accessed measuring its difference phase shift ($\Delta\Phi$) between the two orthogonal basis. It is shown $\Delta\Phi$ is obtained by measuring the rotation angle ($\Delta\theta$) of the elliptical polarization from its initial orientation (θ_0). Considering the effective field phase ($E_{\pm} = A_{\pm} \exp(in_{\pm}k_0z)$) into the eq. (62) and factoring in a convenient way:

$$\mathbf{E} = (A_+ \exp(i\Delta\theta) \hat{\mathbf{e}}_+ + A_- \exp(-i\Delta\theta) \hat{\mathbf{e}}_-) \exp(ik_m z), \quad (73)$$

such as, it is introduced the medium mean propagation wavenumber:

$$k_m = \frac{1}{2}(n_+ + n_-)k_0, \quad (74)$$

and the amplitude phase term:

$$\Delta\theta = \frac{1}{2}k_0\Delta n z. \quad (75)$$

It can be demonstrated that $\Delta\theta$ correspond to a rotation of the semi-axis that defines an elliptical polarization state without changing its eccentricity (minor and major semi-axis ratio).^{79,90-91} Comparing the expression for the rotation angle and the basis phase shift, we see the latter is twice the former. Using eq. (69), we have the NER equation:

$$\Delta\theta = \frac{1}{2}k_0 \left[\frac{B \sin(2\varphi)}{4n_0^2 \epsilon_0 c} \right] LI. \quad (76)$$

As it happens, the B component of $\chi^{(3)}$ may be evaluated measuring the ellipse rotation angle.

3.3.3 From B to n_2

One of the studied effects of $\chi^{(3)}$ is the induced change of the refractive index by the optical field.⁸⁴ The parameter that characterized this change is the nonlinear refractive index (n_2), expressed in the eq. (50). Such is the importance of this parameter, there are plenty of characterization measurements, performed using several techniques, for almost all known materials where it has been observed.⁸⁵⁻⁸⁶

The NER effect allows to obtain the B coefficient of $\chi^{(3)}$ from the ellipse rotation angle, however it is also necessary the value of A in order to compute n_2 (Eq. (70) and (71)). Then, other technique, based in another $\chi^{(3)}$ effect, must be employed to obtain the value of A . Even though, the B to A ratio, well established to ENR and OM, can be used to compute n_2 from solenly the knowledge of one component, thus simplifying the analysis. Substituting the B to A ratio into eq. (70) and (71), also using eq. (50), we get:

$$n_{2,ENR} = \frac{3}{8n_0^2 \epsilon_0 c} B_{ENR} \quad (77)$$

$$n_{2,MO} = \frac{1}{6n_0^2 \epsilon_0 c} B_{MO}, \quad (78)$$

such as the B subscript indicates which ratio was employed. In addition, it should be noticed eq. (48) for χ^* is only valid for a self-induced effect in an isotropic medium using a linear

polarization. Even though, we are using it for an elliptical polarization state, it will be seen the results obtained applying this assumption leads to satisfactory results, even though slight deviations are expected.

The set of eq. (77) and (78) gives the value of n_2 into the two specific cases, pure ENR or saturated MO alignment. Although, it is known ENR contribution is predominant around tens of fs, and from this point on, MO considerably contribute to n_2 up to a complete alignment. Consequently, n_2 is dependent of the beam pulse width (τ_p) and whenever both contributions must to be accounted, eq. (77) and (78) no longer can be used. Keeping that in mind, we define an effective value of n_2 as the sum of both contributions.²³

$$n_{2,eff}(\tau_p) = n_{2,ENR} + n_{2,MO}(\tau_p). \quad (79)$$

Such as the dynamics of ENR process being faster than the employed excitation pulse width, the ENR contribution to $n_{2,eff}$ is assumed as instantaneous, therefore the variation of $n_{2,eff}$ as a function of τ_0 falls onto the MO contribution, which is represented by the function $n_{2,MO}(\tau_p)$ that vary from zero, for a few femtoseconds, to its saturation value, for a few picoseconds.

3.3.4 The MO Proportional Factor

From eq. (78), the same value of B leads into two distinct values of n_2 , depending on the physical mechanism employed.⁸² In our understanding, B values obtained by NER technique are effective values, from which it must be stated with the respective pulse width that induced the effect. Then, from now on, a subscript is added on B and n_2 indicating the regime it was measured. From the ratio of eq. 79 and (78), it is obtained the following relation⁹³:

$$\frac{B_{OM}}{B_{ENR}} = \frac{9}{4} \frac{n_{2,OM}}{n_{2,ENR}}. \quad (80)$$

This factor of 9/4 was experimentally confirmed and explored by Barbano²⁹, demonstrating that a n_2 measurement done by NER are more versatile than Z-Scan ones to measure MO contribution.

3.4 PROPAGATION THROUGH FIBER

3.4.1 The Nonlinear Schrödinger Equation

Now, the Helmholtz equation (eq. (16)) needs to be solved considering the NLO contribution on the dielectric constant, keeping in mind that $n^2(\omega)$ is no longer equal to $\varepsilon(\omega) = 1 + \chi^{(1)}$ (Eq. (53)), because it is intensity dependent. In order to reach the Helmholtz equation from eq. (13), the NLO contribution of eq. (14) should be considered to be very small, which

is justified into the Slowly Varying Envelope Approximation (SVEA) approach. The SVEA prerogative consists in the presumption that in a forward-traveling pulse wave its amplitude envelope varies very slowly in time and space compared to its wavelength.

Employing the method of separation of variables once more, with the exception the electric field amplitude is unrelated of the propagation space coordinate.

$$\tilde{E}_z(r, \omega) = \tilde{A}(z, \omega) F(\rho, \phi) \exp(im\phi) \exp(i\beta_0 z). \quad (81)$$

Then, substituting eq. (81) on (13) and considering the NLO contribution and the SVEA, results into the following coupled differential equations:

$$\left[\frac{\partial^2}{\partial \rho^2} + \frac{1}{\rho} \frac{\partial}{\partial \rho} + \frac{1}{\rho^2} \frac{\partial^2}{\partial \phi^2} + (\varepsilon(\omega)k_0^2 - \tilde{\beta}^2) \right] F(\rho, \phi) = 0, \quad (82)$$

$$\left[2i\beta_0 \frac{\partial}{\partial z} + (\tilde{\beta}^2 - \beta_0) \right] \tilde{A}(z, \omega) = 0, \quad (83)$$

such as $\beta_0 = \beta(\omega_0)$ is the central wavenumber, while $\tilde{\beta}$ is the intensity dependent propagation wavenumber coming from the process of separation of variables. Furthermore, on the eq. (83) the term $\partial^2/\partial z^2$ was neglected due to SVEA.

The intensity dependent wavenumber (Eq. (82)) is solved in the same way as the linear case (eq. (19)). Using a first order perturbation theory ε is replaced by n^2 at a zero order approximation, such as the fundamental distribution mode is obtained. Then, we include the effect of a small variation on refractive index (δn). In the first order, the mode is not affected by δn , but the propagation wavenumber becomes:

$$\tilde{\beta}(\omega) = \beta(\omega) + \Delta\beta(\omega), \quad (84)$$

such as:

$$\Delta\beta = k^2(\omega) \frac{n(\omega)}{\beta(\omega)} \frac{\iint_{-\infty}^{\infty} \delta n |F(\rho, \phi)|^2 d\rho d\phi}{\iint_{-\infty}^{\infty} |F(\rho, \phi)|^2 d\rho d\phi} = \gamma(\omega) |\tilde{A}(z, \omega)|^2 + i \frac{\alpha}{2}, \quad (85)$$

where $\beta(\omega) \approx n(\omega)k(\omega)$; $\delta n = n_2 |\tilde{A}(z, \omega)|$; $\gamma = n_2/k_0 A_{eff}$ is the NLO parameter and A_{eff} is the effective mode area given by:

$$A_{eff} = \frac{\left[\iint_{-\infty}^{\infty} |F(\rho, \phi)|^2 d\rho d\phi \right]^2}{\iint_{-\infty}^{\infty} |F(\rho, \phi)|^4 d\rho d\phi}, \quad (86)$$

The eq. (86) depends only on the fiber geometry, which could be tailored in order to enhance γ . Thus, assuming a fundamental mode propagation and a cylindrical shape for the HCF, $F(\rho, \phi)$ can be approximated by a Gaussian function, and A_{eff} by πw_0^2 . It can be demonstrated that the best coupling is achieved when the ratio w_0/a is:

$$\frac{w_0}{a} \approx 0.65 + 1.619V^{-\frac{3}{2}} + 2.879V^{-6}, \quad (87)$$

which depends on the V number (eq. (26)).

Once the intensity dependent propagation correction has been computed, we can turn our attention to eq. (83). If we assume a quasi-monochromatic pulse, thereby $\Delta\omega \ll \omega$, we can develop the term $(\tilde{\beta}^2 - \beta_0^2)$ into a power series around β_0 , which results in $2\beta_0(\tilde{\beta} - \beta_0)$. The previous approximation states that each spectral component comprising the pulse suffers a phase shift whose magnitude is both frequency and intensity dependent. Furthermore, as the exact function to $\beta(\omega)$ is not usually known, it is developed into a power series around the central wavelength, where the same is done for $\Delta\beta$ in the eq. (85). The series are truncated on the second order for β , and on the zeroth order for $\Delta\beta$, due to the quasi-monochromatic assumption. Considering this, we take the inverse Fourier transform, that brings back the eq. (83) into time domain, reminding the term on $\omega - \omega_0$ are replaced by the differential operator $-(\partial/\partial t)$, then:

$$\left[\frac{\partial}{\partial z} + \frac{i\beta_2}{2} \frac{\partial^2}{\partial t_{gv}^2} - i\gamma(\omega_0)|A|^2 + \frac{\alpha}{2} \right] A = 0, \quad (88)$$

which was written such that the reference moves along the pulse propagation direction at group velocity ($t_{gv} = t - \frac{z}{v_g} = t - \beta_1 z$). The eq. (88) is the widely known as NLO Schrödinger equation (NLSE), and it dictates the behavior of the majority of NLO optical effects linked to the third-order susceptibility in optical fibers. There is also the generalized NLSE, which expand and cover a larger amount of NLO phenomena, but it will not be covered in this work.

3.4.2 Propagation Regimes

The NLSE describe a light pulse propagation through an optical fiber, taking into account its dispersion, NLO effects of $\chi^{(3)}$, and losses. However, one effect may totally overcome by the others, depending on their relative magnitudes, which depends on the initial pulse duration (τ_p) and peak power (PW_0) of the propagating pulse, among others. Dispersion and nonlinearities may compete along the fiber propagation, and in order to highlight the predominance of one effect over the other, we rewrite the eq. (88) introducing two length scales, the dispersion ($L_D = \tau_p^2/|\beta_2|$) and the nonlinear ($L_{NL} = 1/\gamma PW_0$) characteristic lengths. Additionally, it is useful to normalize the time scale and the field amplitude.

$$t_N = t_{gv}/\tau_p, \quad (89)$$

$$A(z, \tau) = \sqrt{PW_0} \exp\left(-\frac{\alpha z}{2}\right) U(z, \tau). \quad (90)$$

Thus, the NLSE is restated as:

$$\left[\frac{\partial}{\partial z} + i \frac{\text{sgn}(\beta_2)}{2L_D} \frac{\partial^2}{\partial t_N^2} - i \frac{\exp(-\alpha z)}{L_{NL}} |U|^2 \right] U = 0, \quad (91)$$

such as $\text{sgn}(\beta_2) = \pm 1$, depending if the propagation is on normal or anomalous regime, respectively.

The length scales L_D and L_{NL} give the length over each effect starts to be relevant to the pulse change along the propagation path. The following cases depict how the relation of those parameters classify the propagation:

- $L_D \gg L$ and $L_{NL} \gg L$: the interaction length is not enough for either effect to take place.
- $L_D < L$ and $L_{NL} \gg L$: only the dispersion will manifest its effects, and the pulse will compress or broaden depending on the sign of β_2 and the pulse chirp.
- $L_D \gg L$ and $L_{NL} < L$: in this scenario, the pulse will be spectrally broadened by SPM.
- $L_D < L$ and $L_{NL} < L$: both effects will influence the pulse shape.

The last case deserves some further analysis. Whenever dispersion and NLO effects act together, the interplay between those effects may lead into a rich variety of phenomena, as soliton propagation and dynamic, pulse self-compression, among others.

3.4.3 Self-Phase Modulation

Self-phase Modulation (SPM) is also a manifestation of a $\chi^{(3)}$ nonlinearity. It is the temporal spectrum analog of the self-focusing effect ($n_2 > 0$), and it can be modeled by neglecting the GVD term in eq. (91), which falls into $L_D \gg L > L_{NL}$ condition. The remaining part of eq. (91) may be solved using $U = V_U \exp(i\psi_{NL})$, such as ψ_{NL} is the NLO phase shift. Equating the real and imaginary part of eq. (91), we have:

$$\frac{\partial V_U}{\partial z} = 0, \quad (92)$$

$$\frac{\partial \psi_{NL}}{\partial z} = \frac{\exp(-\alpha z)}{L_{NL}} V_U^2. \quad (93)$$

Basically, eq. (92) demonstrated SPM phenomena does not affect the pulse shape, while the integration of eq. (93) shows that ψ_{NL} is intensity dependent. The maximum phase shift occurs at the pulse center ($t = 0$), and its value is:

$$\psi_{NL,max} = \frac{L_{eff}}{L_{NL}} = \gamma P W_0 L_{eff}. \quad (94)$$

The SPM creates new spectra components in the propagating pulse due to ψ_{NL} time dependency, thereby introducing new instantaneous frequencies ($\delta\omega$) on the existing pulse spectrum, culminating in an increasing of pulse spectral bandwidth.

$$\delta\omega = -\frac{\partial\psi_{NL}}{\partial t} = -\left(\frac{L_{eff}}{L_{NL}}\right)|U(0,\tau)|, \quad (95)$$

such as $\delta\omega$ is also known by frequency chirping, because it is negative in the leading edge of the pulse (a red shift), and positive in the trailing edge (a blue shift) of the pulse. Thereby, the chirp induced by SPM increases in magnitude as the pulse propagate, which may broaden or shorten the spectrum over its initial state depending on the pulse initial chirp.

3.4.4 Optical Soliton

Whenever GVD and SPM should be taken into account, every term in eq. (91) must be considered, and the relative contribution of both effects is expressed by the soliton order (N_s):

$$N_s^2 = \frac{L_D}{L_{NL}} = \frac{\gamma P_0 \tau_0^2}{|\beta_2|}. \quad (96)$$

The eq. (96) dictates dispersion dominates for values lesser than one, while nonlinearities dominate for values larger than one. However, a fascinating effect happens when both effects compensate each other and N_s value is around one. Whenever it happens, the propagate beam is called an optical soliton, which by definition is a wave packet that can propagate undistorted over long distances. The N_s parameter is also referred as the soliton order.⁶⁶

Specially in the case of gas-filled HCF, where the GVD can be set by the inner gas pressure (eq. (39)) to a suitable value, such as it compensates the increasing of spectra bandwidth added by SPM, a soliton may propagate through the HCF. Figure 4 depicts the numerical solving of eq. (91) for several conditions. Propagation of a sech^2 pulses at 1030 nm with 30 fs duration in a fiber with 50 μm of core diameter filled with 10 bar argon. The left side of Figure 4 depicts the temporal evolution, whereas the right side the spectral evolution. In Figure 4 (a) we see dispersion is the dominant process during the pulse propagation ($N_s=0.1$, 0.7 nJ, $L_D=92$ cm). As the pulse propagates, the dispersion increases τ_0 , but the spectrum of the pulse is not significantly affected. Increasing the pulse energy (67 nJ), SPM effect becomes more and more significant, up to the point the anomalous dispersion is completely compensated by the SPM effect, allowing a soliton ($N_s=1$) to be propagated (Figure 4 (b)). Increasing even further the

pulse energy, there is the formation of a higher order soliton propagation. Figure 4 (c) and (d) are the result for $N_s=2$ (0,27 μJ) and $N_s=3$ (0.61 μJ). We clearly see temporal and spectral wiggles, with period of $(\pi/2)L_D$ ^{66,94} along the propagation. In an initial stage SPM is only partially compensated by the dispersive phase coming from anomalous propagation. Lower-frequency components generated through SPM at the leading edge have a lower group velocity than the center of the pulse. Likewise, the high-frequency generated components at the trailing edge have a higher group velocity. As a result, both are shifted towards the center of the pulse, leading to a temporal self-compression at the distance defined by $\approx L_D/N_s$.⁹⁵

Figure 4 – Simulation of temporal and spectrum evolution of a pulse propagation along the gas filled HCF length.

Source: By KÖTTIG.⁵⁶

3.4.5 Dispersive Wave Emission

The modeling of eq. (91) and the simulation of section 3.4.4 cease to be valid for higher order solitons, whenever few-cycle pulses with one or more octave-spanning spectra comprise the initial pulse. In the previous condition, phase matching to linear waves becomes possible at resonant frequencies away from the soliton central frequency, in the normal dispersion condition, preserving the temporal characteristics of the soliton that originates it, as long as it also maintain the same propagation constant (phase-match condition). This soliton fission can result in a resonant dispersive wave emission (DWE)⁹⁶⁻⁹⁹, which can also be explained by a cascade FWM.¹⁰⁰ In gas-filled HCF, the spectral overlap comes from extreme soliton self-compression of the pulses along the fiber. Figure 5 depict the temporal and spectral dynamics of a DWE propagation along the fiber length. Apart from the octave-spanning spectrum, the conditions of the Figure 5 are the same as Figure 4 (d).

Figure 5 – Simulation of temporal (a) and spectrum (b) evolution of DWE along the fiber propagation.

Source: By KÖTTIG.⁵⁶

Although DWE was already known in SIF, in gas filled HCF it was proposed theoretically in 2010, and experimentally demonstrated in 2011.⁵⁵ The advantages of DWE in gas filled HCF comprises the tuning of the resonant wavelength by the gas pressure⁵⁸, and the fact that it can occur in spectral regions where solid materials absorb light and get damage, limiting the energy of DWE. Additionally, the weak dispersion of noble gases led to a broad phase matching bandwidth, such as ultrashort DWE with few femtoseconds duration can be generated.¹⁰¹

4 CHARACTERIZATION OF GASES NONLINEAR REFRACTIVE INDEX

In this chapter, we review the development of the NER technique and its application on the study of the materials nonlinear refractive index, giving emphasis to gases and their NER signal dependency with the molecular orientation.¹⁰² We recovered how the accuracy of the NER measuring have been improved and used to study the n_2 of vitreous and solvents samples and their comparison with a stablish and standard methods as the Z-Scan. The whole method of measurement NER effect is covered in-depth, along with the setup details, as much for gases as for solid and liquid samples. In the end, the results concerning the gases characterization using the NER effect are presented, where we highlight the study of the effective nonlinear refractive index as a function of the excitation temporal pulse width.

4.1 A NEW WAY OF EVALUATE n_2

The NER effect is employed, in our research group, as a technique to evaluate the materials NLO refractive index.¹⁰³ NER is a stablished technique for measuring n_2 in solid and liquids^{26-28,104-106}, also in the presence of two-photon absorption¹⁰⁷ and having been used even as an innovative tool for imaging microscopy.¹⁰⁸ Our group extended NER measurements into gaseous samples, improved the acquisition method and explored even further its characteristics.¹⁰² A brief description of the latest achievements is given to frame the results present in this work.

As a first approach, standard vitreous samples as silica and BK7, whose n_2 has been vastly measured with several others techniques⁸⁵⁻⁸⁶, were measured using NER and compared with Z-Scan results.¹⁴ The results demonstrate a good agreement with the n_2 value obtained by both techniques²⁶, and several others reported in the literature. Soon after, the NER measurements and model were extended to contemplate thick vitreous samples, demonstrating the possibility to measure more than one sample in a single scan.²⁷

The next step on the technique development consisted on the study of samples with MO contribution to n_2 using NER measurements. The carbon disulfide solution (CS_2) was chosen due to its consecrate NLO properties and its spread use in nonlinear devices. By the time, discrepancies on n_2 values of CS_2 were observed measuring it using the NER technique⁸⁵⁻⁸⁶, which were consistently bigger than those reported on the literature. In order to investigate why samples possessing MO contributions led into discrepant values of n_2 , we came into the

conclusion there is a proportional factor of 9/4 in the NER measurements when performed in MO samples in comparison with ones presenting mainly ENR. We found out this proportional factor was already contemplated by the established theory, even though it was never highlighted before.²⁹ This study have opened the possibility to investigate the intricate relationship among the $\chi^{(3)}$ components and how they are related with each physical mechanism that generate NLO phenomena.

Another difficulty was found on the application of NER measurement to study the n_2 of material that present both ENR and MO contribution. It is not straightforward to distinguish the amount of ENR and MO that contributes to the value of the effective NLO refractive index ($n_{2,eff}$). The literature had already stated the $n_{2,eff}$ dependency on the excitation pulse width (τ_p)¹⁰⁹, but there was a lack of consistent and meticulous measurements. Considering this, we evaluated the $n_{2,eff}$ by NER measurement on samples presenting MO contribution varying τ_0 continuously. The temporal characterization was limited to the range allowed by the compression pair of prisms separation. The results demonstrate that subtle changes on the $n_{2,eff}$ could be measured, changing from pure ENR contribution to the complete MO saturation.^{28,110}

4.2 EXPERIMENTAL SETUP

4.2.1 Excitation Laser

As an excitation source to the NER effect, we have used a Ti:Sapphire amplified system (*Dragon K&M Labs®*) which is capable to deliver a linearly polarized beam, 30 fs pulses at 1 kHz of repetition rate (R_r), 2 mJ of energy with a central wavelength at 790 nm.

We took advantage of the chirped pulse amplification (CPA) system of the laser amplifier to vary controllably the pulse τ_p . By changing the relative distance (Δd) of the compressor pair of gratings, the last stage of the CPA system, from their optimized position (d_0), corresponding a minimum τ_p , we were able to obtain a continuum change of τ_p from tens of fs up to a few ps. Out of the optimized condition, there is a gradual increase of the pulse dispersion (also known as linear chirp) as Δd increases, consequently τ_p becomes longer and longer (Figure 6). Thus, it was necessary evaluate the intra-pulse dispersion influence on the NER effect. However, as it is going to be seen, the dispersion effects were minimum due to the gases low dispersion in the optical range, and then only the temporal broadening of the pulse was taken into consideration.

Figure 6 – Ti:Sapphire amplifier gratings compressor. G – Grating; RM – Retroreflecting Mirror; PM – Plane Mirror; Δd – Relative grating pair distance.

Source: By the author.

The temporal pulse characterization, as a function of Δd , was done by two distinct techniques. A standard and well established auto-correlation (AC) was performed through the SHG of a beta barium borate (BBO) crystal. And, we also explored the own NER measurement to retrieve the pulse τ_p , rising the status of this method into a self-consistent measurement. The NER temporal characterization it is built on the knowledge that noble gases present exclusively ENR contribution, resulting in a constancy of its n_2 value with τ_p . In this way, every change in the NER measurement of a noble gas is a consequence of a change of τ_p , once the pulse fluency is kept invariant (Eq. (97)). Clearly, the absolute n_2 value may be obtained using the pulse parameters (irradiance, energy, temporal width, waist, etc.), interaction length and density. However, this method would lead into intrinsic uncertainties of about 20%, mainly due to average pulse energy evaluation. Then, it is more accurate to use a standard sample to perform a comparative measurement. Fortunately, we can use noble gases as reference samples. The pulse width and energy can be retrieved from a noble gas NER signal since its value of n_2 is well known. On practical aspects, $n_{2,ENR}$ of a noble gas leads to a respective value of B_{ENR} (Eq. (77)), then through eq. (76) it is computed the pulse intensity (I) (eq. (97) assuming a temporal and spatial gaussian profiles).

$$I_0 = \frac{4\sqrt{\ln 2}PW}{R_r\pi^{3/2}w_0^2\tau_p}. \quad (97)$$

such as PW is the temporal average power of light coupled on HCF. The coupled light power was estimated measuring the output light power coming out of the HCF. The beam waist was chosen to be 64% of the HCF inner diameter (eq. 87).

In this work, we have chosen argon (Ar) as a reference sample ($n_2=10\times10^{-24}$ m²/W, $B\approx7.08\times10^{-26}$ m²/V²).²³. Then using the standard sample, eq. (77), (79) and (97), we have obtained the value of τ_p for value of Δd .

4.2.2 Measuring NER

The initial elliptical polarization state is generated introducing a quarter wavelength plate ($\lambda/4$) on the excitation beam path. The eccentricity of the polarization is defined by the angle the fast axis of the quarter waveplate makes with the linear polarized excitation beam, which coincides with the one defined in eq. (68). When $0 < \varphi < \pi/4$ the polarization state is elliptical, and suitable for inducing the NER effect.

Once set the polarization state of the beam, it is focused on the sample by a convergent lens ($f=30$ cm). The beam interacts with the sample differently whether the sample is solid/liquid or gaseous, and sections 4.2.3 and 4.2.4 target on each one individually. After passing through the sample, a wire-grid analyzer, appropriated for high energy beams, probes the final polarization state. The signal is acquired by focusing the beam with convergent lens in a pin silicon photodiode, whose signal, proportional to the light intensity, goes to an amplifier and is recorded via computer.

The novelty introduced by us on measurement of the NER signal makes its acquisition faster and more sensitive than the one performed previously.⁸⁰ Our acquisition method consists on the usage of a homemade motorized support that spins the analyzer in a given frequency of tens of revolutions per second, henceforth called as signal modulator. The signal modulator also produces an electronic signal with the same frequency of the spinning, analyzing it to a dual-phase lock-in amplifier. The spinning analyzer modulates the light with a controllable frequency, thus creating an envelope for the pulsed signals. A dual-phase lock-in filters and amplifies the electrical signal coming from the photodetector, and synchronizes it with the one coming from the signal modulator. Then, only the light signal with the same frequency of the modulation is amplified and recorded, turning its acquisition very sensitive for reducing vastly the measurement noise.

The major advantage of the dual-phase lock-in is to measure precisely phase shifts ($\Delta\Psi$). Thus, in order to take advantage of that characteristic, we had established a reference modulation, such as we know its relative phase value (Ψ_0), that must be independent of the

NER signal. As usual, we have chosen the reference phase value to be the one defined by the modulation when there is any NLO contribution (Figure 6 (b)), in other words, when light intensity is in the linear regime. Then, introducing the sample on the beam path, or increasing the light intensity up to the NLO regime, the ellipse axis rotation due to NER effect it is going to make the modulation suffer a phase shift from the reference value ($\Delta\Psi$) (Figure 6 (d)), which is sensitively detected by the dual-phase lock-in. It happens the envelope phase shift is twice the value of the ellipse axis rotation angle ($\Delta\Psi = 2\Delta\theta$) according to the Malus' law.⁶⁴ This method is sensitive enough to detect rotation angles smaller than one degree.

Figure 7 depicts the modulation signal for cases where the analyzer impinging light have a polarization state linear (a), elliptic (b) and circular (c). The Figure 7 (d) is the representation of the modulation phase shift due to the elliptical rotation NER effect.

Figure 7 – Envelope modulation signal considering pulses with polarization: (a) linear; (b) elliptical; (c) circular, (d) elliptical after an axis rotation.

Source – By the author.

In the linear case (Figure 7 (a)), no phase shift is expected, because a linear polarization do not suffer an axis rotation through the NER effect. In counterpart with the circular case (Figure 7 (c)), where the phase shift is maximum, though it cannot be measured, because the reference axis cannot be defined.

4.2.3 Liquid and Solid Samples

For liquid and solid samples, the NER measurement is performed scanning the sample along the propagation axis of a focused excitation beam. The NLO effect manifests itself only on the Rayleigh range, where light intensity is higher and the wavefront is plane. Then, far from the focal position, where the intensity is low and only linear optical effects take place, the reference modulation phase shift is defined. As the sample goes towards the focus, the intensity gets higher, and the NER effect starts to manifest. The modulation phase shift reaches its maximum value when the sample is at, or encompasses the Rayleigh range. After focus the signal is symmetric, it gets lower and lower, until returning to the reference phase another time. A representation of the experimental setup is depicted in Figure 8.

Figure 8 – NER experimental setup used for liquid and solid samples. Side (a) and perspective (b) views. WP – quarter waveplate; L – Lens; SH – Sample Holder; M – Modulator; D – Silicon Pin Detector.

Source: By the author.

This measurement method has the advantage that two samples can be evaluated together, at the same scan, as much as the thickness of each sample is larger than z_R . This allows the measurement of the n_2 ratio of samples, giving a reliable comparison. Once a reference sample is chosen, other unknown samples may be scanned together and compared. The difference in amplitude readily gives the proportion of their signals, consequently, their NLO magnitude. For instance, a quartz cuvette is often used as a sample holder for liquid samples, and it provides a reference scale for the nonlinearity of the liquid. Therefore, no evaluation of the optical power is needed, which decreases considerably the uncertainty of the measurement.

4.2.4 Gaseous Samples

The experimental setup used for the NER measurement in gaseous media is similar to the one employed for solid and liquid samples.²⁶ Solid and liquid with NLO properties have a much higher value of n_2 than gaseous media, and an interaction length of order of a Rayleigh range of a focused beam is enough to the NLO signal increase into a measureable quantity. However, for the NER measurement of gas samples, due to their much weaker NLO signal, a HCF propagation is necessary in order to increase the interaction length, scaling up the NER signal.¹¹¹ Consequently, a HCF made of fused silica was used with proper dimensions in order to increase the light propagation. Filling the HCF with a gas, the light-sample interaction in NLO conditions is extended to tens of centimeters with a considerable low loss. The light coupling into the HCF was performed using three-dimensional translational stages. They were attached at HCF tips and adjusted such as its output light was maximized, while keeping the propagation mode Gaussian in the HCF exit. Additionally, the HCF dimensions (150 μm and 1.2 mm of internal and external diameters) were chosen to minimize the losses. The HCF length varied from 20 up to 50 cm, in such a way the light intensity along the internal propagation were considered constant.

The HCF straightness and rigidity was guaranteed by a cylindrical glass matrix, with an internal hole large enough just to fit the HCF (1.22 mm). The external diameter of the glass matrix (8 mm) was chosen to fit the vacuum/injection atmosphere control system. Metallic tubes with vacuum connectors (Swagelok®) in one side, and Brewster angle 0.5 mm quartz windows in the other, were used to seal the glass matrix and control its internal atmosphere. The metallic tubes were connected to a vacuum pump and into a gas injection system. The windows were far enough, 20 cm from the HCF tips, to preventing any NLO effect on them.

The atmosphere control system allowed the gas be confined inside of the HCF with a controllable pressure, from 10^{-2} Torr up to one atmosphere.

Differently from the solid/liquid setup, the reference modulation phase signal was obtained evacuating the HCF system into a pressure where the NLO effects are negligible, due to the gas extreme low density. Rising the gas pressure, the NER effect increases and it is recorded when it reaches 1 atm. The gaseous NER experimental setup is depicted in the Figure 9.

Figure 9 – NER experimental setup used for gaseous samples. Side (a) and perspective (b) views. WP – quarter waveplate; L – Lens; SH – Sample Holder; GF – Gas Flow; M – Modulator; D – Silicon Pin Detector.
Source: By the author.

4.3 NER ON GASES

4.3.1 Parameters Characterization

In this chapter, we describe the experimental results, in which several parameters were explored with the aim of a better understanding of the gaseous NER measurements setup. We have taken a set of eleven full acquisitions of all samples, varying τ_p in the whole range and

performing a NER temporal calibration for each set of measurement. Between the sets, we have changed the incident pulse energy, trying to avoid any influence of other effect or saturation, while preserving the signal ratio among the samples. We also vary the HCF length, in order to better know the hole of dispersion and loss on the system. The parameter of each set is depicted in Table 1.

Table 1 – Nomenclature and parameters of each set of NER measurements.

Set	L (cm)	E_p (μ J)	Set	L (cm)	E_p (μ J)
#1	20	10	#7	20	30 for Ar 13 for N ₂ , Air and O ₂
#2	20	10	#8	30	14 for Ar 11 for N ₂ , Air and O ₂
#3	20	20	#9	40	11 for Ar 8 for N ₂ , Air and O ₂
#4	20	13	#10	50	20 for Ar and He 1 for N ₂ 8 for Air 10 for O ₂
#5	20	40	#11	50	22 for Ar 8 for N ₂ , Air and O ₂
#6	20	15			

Source: By the author.

In the sense of completeness, we have characterized other parameters related with the gaseous NER technique. We measured the gaseous NER effect as a function of the polarization state (Figure 10 (a)), varying the quarter-waveplate fast-axis angle relatively to the incident linear polarization (φ). As described in eq. (76), $\Delta\theta$ is zero for $\varphi = 0^\circ$ and 90° (linear polarization), and undefined for $\varphi = 45^\circ$ (circular polarization) (Eq. (66)). Any other value of φ can be used to produce the necessary elliptical polarization and, consequently, to compute the B component. However, we have chosen to use $\varphi=25^\circ$ for all measurements, because it is an intermediate angle between linear and circular polarization state, and gives the better signal to noise ration. Thus, we have also explored the NER signal as a function of the gas pressure (Figure 10 (b)) and the coupled pulse energy in the HCF (Figure 10 (c)).

As expected, Figure 10 (b) shows a linear behavior in the measured range.^{17,111} For improving even further the accuracy of the NER measurement, the rotation angle at a given pressure was determined by a linear fit of a pressure ramp. Thus, from now on, all measurements of $\Delta\theta$ are given at the local atmospheric pressure (690 Torr). Furthermore, Figure 10 (b) shows the amplitude of the effect is increasingly larger for Ar, N₂, Air and O₂, and it goes along with their respective densities, ionization potential, refractive index, and others. The even weaker signal of noble gasses attributed to the fact it possesses a close electronic shell, which makes harder to induce a NLO polarization. Additionally, we collected the NER signal for several

values of pulse energy, showing a linear tendency, also as expected by eq. (76). Figure 10 (c) shows $\Delta\theta$, for a fixed elliptical polarization state, as a function of the mean throughput pulse energy ($\bar{E}_p$) coupled on the HCF.

Figure 10 – NER dependency as a function of: (a) initial polarization state; (b) HCF gas pressure; (c) pulse energy couple into HCF HCF with Ti:Sapphire laser at 800 nm and ~ 60 fs pulse width. In (b) and (c) the NER signal were obtained with polarization state set at $\phi=22.5$ degree.

Source: By the author.

We have observed for $\bar{E}_p$ values above $30 \mu\text{W}$, or when $\Delta\theta$ is above 30° , than the NER signal is no longer linear with $\bar{E}_p$ the gas pressure. Shortly above this threshold, the NER signals tend to saturate, reaching into a plateau. Well above, we have observed a decrease of the signal. The behavior off the linear regime, deserve further investigation, for the understanding of the physical phenomena behind the nonlinearity of those parameters.

Once having the NER measurement basic parameters and their limits well understood, we set out to characterize the NER signal varying the temporal pulse width of the excitation beam. We followed the same procedure for all measurements. Once we have set a particular distance Δd of the compression gratings, thus fixing τ_p , we measure the NER signal for all gases in a row, performing a pressure ramp and recording their θ value at 690 Torr. A change of

Δd in 0.5mm is performed, then we repeat the same procedure up to the point the entire Δd range was swept (28 mm).

4.3.2 Compressor Temporal Calibration

Nowadays, the auto-correlation (AC) method or the frequency resolved optical gating (FROG) are the vastly employed technique used to evaluate short light pulse width.¹¹² However, we noticed the NER signal of a noble gas could be used to obtain a temporal pulse width calibration of the compressor. The Ar NER calibration can be done by substituting the literature value of $n_{2,ENR}$ for Ar ($1 \times 10^{-23} \text{ m}^2/\text{W}$)^{16,21,23,86,111,113} (or measuring its NER signal along with an AC measurement) in eq. (77), converting it into B_{ENR} , and then using eq. (76) and (97) to retrieve τ_p . As the B_{ENR} for a noble gas is assumed constant for pulses longer than femtosecond, the variation in $\Delta\theta$ with Δd is exclusively due to a change in τ_p , when the fluence is kept constant.

We have observed the temperature and the HCF length can change the calibration. The room temperature influences mainly the alignment of the pump laser, and for a long HCF propagation, the gas medium dispersion may become relevant. Then, performing an Ar NER measurement, we could calibrate temporally the compressor for every set of measurement, without any additional AC measurement, assuring a better correspondence of the results.

In order to demonstrate the possibility of using the Ar NER signal as way to determine τ_p , we compared its measurements by the AR NER and AC measurement. Figure 11 shows the set up built and used by us, where the nonlinear effect used for the AC measurement was SHG of a Beta Barium Borate (BBO) crystal.

Figure 11 – Auto-Correlator setup. BS – Beam Splitter; PM – Plane Mirror; CM – Concave Mirror; NLM – NLO Medium; L – Lens; D – Silicon Pin Detector; TD – Temporal Delay.
Source: By the author.

The calibration of the pulse width with Δd using auto-correlation is depicted in Figure 12.

Figure 12 – (a) Calibration of the excitation beam compressor pair of diffraction gratings by a SHG auto-correlation of the excitation pulse; (b) zoom of the shortest pulse region.

Source: By the author.

It can be seen in Figure 12, τ_p that can be tuned continuously from (b) 40 fs to (a) 1.3 ps. Although the minimum pulse width allowed by the fourier transform limit (FTL) of the laser spectrum bandwidth source is 30 fs, any compressor misaligning and/or optics elements propagation before the entrance of the HCF may stretch the incident pulse from its optimum condition.

Figure 13 compares temporal calibration made by SHG AC and Ar NER using several HCF length.

Figure 13 – Comparison between AC and Ar NER temporal calibration for several HCF length.
Fonte: By the author.

The conjecture is that the deviation of τ_p from the SHG AC values for $L = 40$ and 50 cm is probably due to gas dispersion along the HCF propagation. We see τ_p broadens by a few picoseconds on the region of minimum compression, meanwhile it is practically the same for the shortest pulse. The calibration by Ar NER measurement using a 20 cm HCF is practically the same as the one measured by SHG AC, which corroborate the assumption of the noble gas constant NLO effect in the range studied.

4.4 GASES EFFECTIVE n_2

Several NER measurements were performed for all gases on this study, along with their respective temporal calibrations, whereupon we computed the samples B_{eff} coefficient. The result of all sets of measurements can be visualized on Figure 14 for N_2 (a), Air (b) and O_2 (c).

Figure 14 – Effective B value for all sets of measurements as a function of τ_p for: (a) N_2 , (b) Air and (c) O_2
Source: By the author.

The B_{eff} of the molecular gases increase with the pulse width up to reach into a plateau, representing the majority of molecules alignment and a saturation of the MO contribution. It is known that for even shorter pulse width another plateau should be reached, representing exclusively the ENR contribution. The shortest pulse achieved by the laser system was not enough to reach into the ENR plateau, and an additional spectrum broadening followed by a compression would be necessary to reach this temporal range.

It is understood that after a quick ENR response to the optical field, MO contribution start to saturate around 150 fs^{23} , which is corroborated by the data in Figure 14 due to the fast B_{eff} growing, followed by a slower transition to the MO saturation.¹¹⁴

The error associated with B_{eff} is due to the sensibility to the experimental condition, for small room temperature variation, which influences on the operation of the excitation laser system. However, the measurement #11 clearly stands out from the others in the Figure 14, for presenting a mean value different from the whole set, and it was also the one that reaches the biggest temporal range. Based on that, we have chosen to adopt the #11 measurement as the representative over all others. The Figure 15 depicts a cluster of B_{eff} values for all samples.

Figure 15 – Effective #11 B values for all gases, including He.
Source: By the author.

The sensitivity of our NER measurement is bespoken by the capability of measuring the He value of n_2 , whose magnitude is even lower than the one for Ar, thereby its characterization is almost absent in literature. Notwithstanding, the He value was only possible to measure very close of the shortest τ_p values, where the intensity is the highest possible. Besides, He measurements were made together with Ar to ensure the temporal calibration is the same, consequently, limiting the input intensity to hundreds of μJ in order to avoid spurious effects on the other gases measurements. The $B_{eff}(\text{He})$ values showed to be constant using the Ar temporal calibration, which is another indicative of the validity of assumption that noble gases constancy of n_2 on the τ_p range studied. Calculating $n_{2,ENR}$ for He, we have obtained the value of $0.3 \cdot 10^{-24} \text{ m}^2/W$, in agreement with the one reported literature.^{16,115}

At the moment, there is no analytical expression based on theory for modeling the $n_{2,eff}$ dependency with τ_p , consequently, we propose an empirical model (eq. (98)) to fit the results exhibited in Figure 15, which is inspired on an exponential growth. Our model describes that an initial value of the B_{eff} come from a pure ENR contribution, growing up to reach a MO saturation, contemplating a characteristic time growth parameter (τ_g).

$$B_{eff} = B_{ENR} + B_{MO} \left(1 - e^{-\frac{\tau_p}{\tau_g}} \right). \quad (98)$$

Using the model, we fit Figure 15 data, letting three free parameters B_{ENR} , B_{MO} and τ_g , the B values for ENR, MO and the growth parameter, respectively. The fit is depicted in Figure 16.

Figure 16 – Fitting of the #11 set of measurements using the empiric model of eq. (98)
Source: By the author.

The parameters of the fitting are shown in Table 2.

Table 2 - Figure 16 data fit parameters.

Sample	$B_{ENR}(10^{-26}m^2/V^2)$	$B_{OM}(10^{-26}m^2/V^2)$	$\tau_g(\text{ps})$
He	0.2	-	-
Ar	7.1	-	-
N ₂	5.5	47	0.28
Air	5.6	62	0.45
O ₂	5.7	127	0.46

Source: By the author.

Once the values of B_{ENR} and B_{MO} have been determined, it is straightforward to obtain the samples values of $n_{2,ENR}$ and $n_{2,MO}$ using the eq. (78) and (79) (Table 3).

Analogously to eq. (98), we can use the eq. (78) and (79) to write:

$$n_{eff} = n_{ENR} + n_{MO} \left(1 - e^{-\frac{\tau_p}{\tau_g}} \right). \quad (99)$$

Now, eq. (99) and τ_g give us the possibility to compute $n_{2,eff}$ for any value of τ_p in the range of transition between ENR and MO contributions. Then, we can compare our results, using the values on Table 3 and the eq. (99), with the ones reported in the literature (Table 4).¹⁶⁻

Table 3 - $n_{2,ENR}$ and $n_{2,MO}$ values of Table 2 converted by eq. (78).

Sample	$n_{2,ENR}(10^{-24}m^2/W)$	$n_{2,MO}(10^{-24}m^2/W)$
He	0,3	-
Ar	10	-
N ₂	7.8	30
Air	7.9	39
O ₂	8.1	80

Source: By the author.

The table 4 show the values of the effective n_2 (Eq. 80) characterized by other techniques present in the literature. Considering the respective τ_p employed in each experiment, our results agree with the ones reported. Nevertheless, as mentioned before, the fact that our limited pulse width is larger than the one needed to obtain the ENR plateau, letting the value of B_{ENR} free on the eq. (98), probably B_{ENR} is likely to be overestimated, and that may be the reason for the differences reported.

Table 4 - Comparison of the literature values for the $n_{2,eff}(10^{-24}m^2/W)$ with the ones reported in this work.

Sample	Ref. 16 $\tau_p \sim 120$ fs	Ref. 23 $\tau_p \sim 150$ fs	Ref. 16 $\tau_p \sim$ ns	This Work $\tau_p \sim 1$ ps
N ₂	23	32	45	38
Air	29	-	55	47
O ₂	51	63	97	88

Source: By the author.

Although our model provides a characteristic growth parameter for the OM contribution, it should not be confused with the relaxation time of the gaseous molecules. In order to measure such parameter, a single beam technique is not enough, and experiments as pump-probe employing two beams, along with a relative controllable temporal delay, must be performed to access such parameter. Nevertheless, τ_g gives some information regarding the molecules relaxation time, which is not often found in the literature. Then, we believe the τ_g values found by the NER measuring may contribute to bring some light into future experimental approaches.

We also figured that the B_{eff} values for the atmospheric constituents follows their respective mass proportion in the values measured for Air. Into the atmosphere composition, there are approximately 0.01% of Ar, 0.78% of N₂ and 0.21% of O₂. Then considering the saturated values of $n_{2,eff} (= n_{2,ENR} + n_{2,OM})$, their summation weighed by its mass

contribution gives the precise value of Air ($n_{2,ef}(\text{Air}) = 0.01n_{2,ef}(\text{Ar}) + 0.78n_{2,ef}(\text{N}_2) + 0.21n_{2,ef}(\text{O}_2) \cong 45 \times 10^{-24}m^2/W$). That characteristic may lead into a proportion detection of a known gas in a mixture by the NER measurement.

5 ENERGETIC DEEP UV GENERATION

As a first approach to present the DUV generation, we describe how we have compressed the output of a double amplified Ti:Sapphire laser, broadened into an octave, in a sub-10 fs infrared (IR) pulse. The setting of the proper experimental conditions required to excite the dispersive wave emission, resulting from a high order soliton fission. And then, the isolation and characterization of the DUV generation. All the experimental details and setup are rigorously exposed, concerning the sub-10 fs pulse compression, the DUV generation and the temporal characterization.

5.1 IR SUB-10 fs

The generation of DUV by DWE with desired properties demands a high intensity (hundreds of μJ) and short (few fs) IR excitation source, therefore an octave spanning spectrum bandwidth is necessary. In order to produce such a source, we started from a double amplified Ti:Sapphire laser, which was spectrum broadened and posteriorly compressed into a sub-10 fs. In the following sections, we explain in detail each step of the IR sub-10 fs generation, along with the proper characterization.

5.1.1 Excitation Laser

In order to the dispersive wave be emitted into the deep ultraviolet (DUV) range, the soliton propagation dispersion and self-compression must be induced by a high energetic ($\sim\text{mJ}$) sub-10 fs excitation pulse. Previous measurements⁵⁹ showed up that to achieve the desired energy and temporal pulse width, the excitation beam must have around 1 mJ and sub-10 fs, at 790 nm. For the generation of such excitation pulse, we employed a double amplified Ti:Sapphire system (15 mJ, 1 kHz, 45 fs), which was spectrally broadened by SPM for latter compression. Temporal characterization of the primary excitation beam was performed by a home-built SHG-FROG. The spectrogram is depicted in Figure 17.

Figure 17 – SHG-FROG (a) experimental and (b) retrieved spectrograms of the primary excitation pulse; (c) normalized SC trace and the retrieved pulse intensity; normalized spectrum and retrieved phase.
Source: By the author.

The genetic algorithm used for the retrieval of the spectrogram reveals a 45 fs, close from que FTL of laser spectral bandwidth (22.6 nm). The beam energy had to be adjusted, such as after the compression the pulse energy was a few mJ. Therefore, an IR beam-splitter was placed just after the laser output, deviating 2.5 mJ to be coupled on a 1 m long HCF fiber filled with Ne for SPM broadening. The 2 cm beam diameter was focused by a convergent silver mirror into the SPM-HCF.

5.1.2 Spectrum Broadening

For spectrum broadening, 2.5 mJ of the excitation beam were coupled into a silica HCF (ID=250 μm , ED=350 μm , L = 1 m) filled with Ne by a concave silver mirror (f=1250 mm). The mirror focal length was chosen to maximize the mode coupling, which is achievable by choosing the focusing length such as its w_0 match 64% of HFC internal diameter. The whole system is confined inside of a metal KF-40 tube, dubbed SPM-housing, to achieve a Ne pressure control. Both tube endings had Brewster fused silica windows (SiO_2 – 190 -20000 nm) of 0.5 mm thickness, and 30 cm apart from the fiber endings to avoid any NLO effect on them (Figure 18). The transmission output of the system, after gas evacuation, was measured to be 55%.

Figure 18 – SPM HCF Housing. BW – Brewster Window; GF – Gas Flow; VG – V-Groove.
Source: By the author.

As soon as Ne was injected into the SPM-housing, the phenomena of SPM take place. The noble gases are chosen, as NLO media, for they weak dispersion, which enhance the SPM efficiently. Furthermore, the high ionization threshold of noble gases are beneficial when milli-Joules pulse propagation is considered. Specifically, neon (Ne) was chosen due to its high ratio of n_2 ($6 \times 10^{-25} \text{ m}^2/\text{W}$) and its ionization potential (2080 kJ/mol), and also for its availability in comparison with krypton (Kr) e xenon (Xe), that are more costly. Figure 19 exhibit the spectral broadening for five distinct Ne pressure values.

Figure 19 – SPM-HCF output spectrum for several values of Ne filling pressure.
Source: By the author.

Even at a very small quantity of Ne is enough to generate spectral broadening. It reaches into a saturation of the effect at 1.2 bar, in such a way the final spectrum is comprehend from 570 nm up to a distance IR ($>1 \mu\text{m}$). The efficiency of the spectrometer falls drastically after 1 μm , and the acquired spectra were not corrected.

5.1.3 Pulse Compression

After emerging from the SPM-housing the beam is collimated and steered into a battery of eight double angle dielectric chirped mirrors (CM) (Figure 21), each one introducing a fix amount of negative second order dispersion per bounce ($\beta_2 = -40 \text{ fs}^2$).

Figure 20 – Chirped mirror compressor, double 5° and 19° architecture.

Source: By the author.

The chirped mirrors spectral range covered from 500 to 1050 nm, centered at 775 nm. A double angle (5° e 19°) architecture guarantees a more efficient dispersion introduction control, avoiding the oscillations present in single angle architecture, intrinsic from their manufacture process (Ultrafast Innovations®, PC70 - Figure 20). On plus, we put a pair of quartz wedges (10 mm of maximum thickness) on the beam path after the mirrors, adding positive dispersion in a controllable way, in order fine tune the dispersion introduced on the pulse. The wedges are crucial to compensate any extra dispersion introduced by the CM, or any other optical components, before reaching into the DWE-HCF.

Figure 21 – Second order dispersion and reflectivity spectrum of the PC70.
Source: By Ultrafast Innovations GmbH.

After the compression system, the emerging pulse (0.9 mJ) was evaluated by several FROG measurements. We experiment several wedges thickness configurations to determine the shortest pulse possible to achieve. Due to the large pulse spectral bandwidth after the SPM-HCF, we have chosen to use SD instead of SHG as NLO effect for the FROG measurements.^{34,116} The FROG measurements setup was composed by a pair of plane D-shaped mirrors, which generate to replicas of the probed beam. One of the D-shaped mirrors was mounted onto a linear translator, in order to provide the needed temporal shift between the two copies. The replicas should be close of each other, such as they could interact in the NLO medium with a very small angle ($\sim 2^\circ$). A concave mirror with 10 cm of focal length focus the beam in the NLO medium. Then, the spectrum of the signal is collected as a function of the temporal overlap between the two replicas. As NLO medium, we chose a Sapphire crystal with 100 μm thickness for the SD-FROG (Figure 22).

Figure 22 – SD-FROG setup. DM – D-Shaped Mirror; CM – Concave Mirror; NLM – NLO Medium; SD – Self-Diffraction Signal; D – Silicon Pin Detector; TD – Temporal Delay.
Source: By the Author.

The SD-FROG spectrogram of the shortest pulse is depicted in Figure 23.

Figure 23 – SD-FROG (a) experimental and (b) retrieved spectrograms of the sub-10 fs excitation pulse; (c) normalized AC trace and the retrieved pulse intensity; normalized spectrum and retrieved phase.
Source: By the author.

Once we achieve the desired pulse width employing SD-FROG measurements, we performed SHG-FROG, with the same setup, only exchanging the sapphire by a 10 μm thick BBO. The Figure 24 shows the experimental and retrieved spectrograms obtained by the SHG FROG.

Figure 24 – SHG-FROG (a) experimental and (b) retrieved spectrograms of the sub-10 fs excitation pulse; (c) normalized AC trace and the retrieved pulse intensity; normalized spectrum and retrieved phase.

Source: By the author.

The agreement in the temporal pulse width using both effects as NLO effect, corroborate the measured sub-10 fs pulse obtained, which is suitable as initial conditions to generate the short DUV by DWE.

5.2 DWE SETUP

After losses on the pulse compression, 0.8 mJ of pulse energy was coupled into the DWE-HCF, using a 50 cm focal length focusing silver mirror. Once more, the focal length was chosen, as before, to maximize the mode coupling. The DWE setup is comprised by the stretched flexible HCF (Micro Quartz® ID=250 μm , ED= 360 μm , L =2 m) and its housing, support, attached to the vacuum parts for atmosphere control, and entrance and output optical

windows. Calcium fluoride (CaF_2) windows, 0.5 mm thickness, 30 apart from the fiber tips, at Brewster angle seal the vacuum system and avoid absorption of the generated DUV beam.

The silica HCF length limitation has been suppressed by the development of the flexible HCF, which was demonstrated to work without huge losses up to 5 m. Nonetheless, it was necessary to stretch the fiber, applying a certain amount of linear tension, in order to guarantee proper straightness. Stretching the flexible HCF demands a mechanical system that pulls both fiber tips without any bending and twisting, while keeping it in vacuum. It is highly desirable to be able to control the fiber stretching and positioning translators on the fiber tips, because the energy and the spatial mode that comes out are very sensitive to both of them. Despite the fact there are commercial opto-mechanic parts available to stretch flexible HCF (Few-Cycle® Fiber), they are very costly. Then, we have decided to manufacture our own fiber stretcher, based on the original ideas of Tomas Nagy.⁴⁷

The condition whereby we have assessed the straightness of the flexible HCF was based on the throughput power and mode of a couple He-Ne laser beam. We managed to couple 70% of He-Ne energy throughout the 2 m flexible HCF, along with a good fundamental mode. However, when we coupled the IR sub-10 fs excitation beam, only 30% passed through the same system, much lower than the 60 to 70% expected. The spatial mode of the excitation beam suffered modifications and it was no longer the fundamental mode. Even though SFM-HCF acted as a spatial filter, defects on its output tip and the several bouncing on dispersive (a.k.a. chirped) mirrors compressor may have caused deviation of the spatial profile. Additionally, it is known the losses scale with the pulse energy.

5.2.1 Propagation characterization

In order of characterize the spectral changes along the whole system, we acquired the output spectrum after the DWE-HCF in several conditions. First, we have observed the spectral changes as a function of the SPM-HCF Ne filling pressure, keeping the DWE-HCF evacuated (Figure 25 (a)). It is known, for a temporally symmetric excitation pulse, that we would also expect the SPM broadening to be symmetric. Despite this fact, Figure 19 shows an asymmetric broadening, which might be an indication of temporal changes in the pulse envelop consequence of the dispersion introduced at the CM assembly. In a second scenario, we have the SPM-HCF pressure fixed in 1.2 bar and increased the DWE-HCF Ne pressure (Figure 25 (b)). Figure 25 shows a more symmetric/homogeneous SPM when we increase the DWE-HCF gas pressure. The throughput beam became brighter and brighter with the increasing pressure, without any change the pulse energy, an indication of a super continuum generation.

Figure 25 – DWE-HCF output spectrum for several values of Ne filling pressure when (a) SPM-HCF is evacuated and (b) filled with 1.2 bar of Ne.

Source: By the author.

5.3 DUV GENERATION

5.3.1 Spectrum Characterization

The generated DUV coming out of the DWE system is concentric with the pump excitation beam. In a first attempt to isolate the DUV from the excitation beam, a couple of UV dielectric mirrors, centered at 250 nm and with enough bandwidth work range, were employed to filter the excitation beam. However, they were not able to remove the entire excitation beam and, consequently, the efficiency of the process could not be evaluated, but the spectrum of the throughput beam showed clearly the energy transfer to the DUV region (Figure 26).

Figure 26 – Spectrum of the DWE transfer of energy into the DUV region.

Source: By the author.

In the Figure 26 we observe that DWE has a peak at 225 nm and there is a clear separation region between it and the excitation beam spectrum, exactly where a second order zero dispersion is predicted ($\lambda_{zd} \cong 400 \text{ nm}$). Even though DUV generation is clear, we had to confirm by which process the DUV were produced. Some NLO process like THG or FWM, could generate the DUV. However, the pressure tuning λ_{zd} was used to solve the ambiguity about which phenomena generates the DUV lays on a DWE property. The central wavelength of DWE can be tuned by the gas pressure in DWE-HCF, excitation energy or temporal width of the excitation pulse, which does not occur on the others NLO mechanism.

In order to proper filtering the DUV, we steered the outcome of DWE system to pass through a magnesium fluoride (MgF_2) prism, which is known by its good DUV transparency property, and made the separation by dispersion (Figure 27).

Figure 27 – Scheme of the DWE separation from the pump beam. PM – Aluminum Plane Mirror; P – MgF_2 Prism; S – Stopper; SP – Spectrum Photometer.

Source: By the author.

Figure 28 shows a picture of the beam impinging a white paper after the prism.

Figure 28 – Picture of DWE beam after passing through a MgF_2 prism, projected in a white paper.
Source: By the author.

The right most beam on the Figure 28 is the remaining of the excitation beam, while the three others to its left correspond to DWE. The Figure 29 depicts the spectrum of all three DWE beam presented in Figure 28.

Figure 29 – DUV beam spectra after a MgF_2 prism.
Source: By the author.

We highlight that in Figure 28 there is a fourth beam around 200 nm, though it could be barely recorded because it is quite on the edge of the used spectrometer range. We had to use a couple of quartz plates reflections, in order to decreasing the DUV fluency, to focus the beam into the spectrometer.

The confirmation of the DUV generation by DWE was realized acquiring the spectrum of the filtered beam for some DWE-HCF pressure values. The result is shown in Figure 30 along with the average power measurements.

Figure 30 – DUV spectrum as a function of DWE-HCF Ne filling pressure.
Source: By the author.

As we can see in Figure 30, the central DWE wavelength suffers a red shift with the increasing DWE-HCF pressure, then confirming the DWE pressure dependency, with energy value enough to induce NLO processes. However, for the lowest pressure (≈ 0.2 bar), the first shift occurs towards the extreme UV, and just after it goes to longer wavelengths. We have found no references that could explain such behavior, which deserve a further investigation.

Furthermore, we explore the DWE for other noble gases as NLO media. The results for Ar and He are exhibited in Figure 31.

Figure 31 – DUV spectrum as a function of HCF (a) Ar and (b) He filling pressure.
Source: By the author.

The Ar and He present the same behavior as Ne, considering the DWE properties, with the exception of the pressure shift magnitude and energy. It is known Ar has a larger and He a lower $n_{2,ENR}$ than Ne, consequently Ar reaches 270 nm in 0.3 bar, whereas He only gets 250 nm using 0.8 bar. As DWE is related with a SPM process, we relate the behavior of the other gases with its ionization potential and their NLO index of refraction, because the larger the NLO medium $n_{2,ENR}$, the bigger is the rate of central DWE wavelength shift with gas pressure.

5.3.2 Temporal Characterization

At last, once we could get the DUV with the desired energy and tunability, it was necessary to temporally characterize the DWE. According to numerical simulations^{56,95} the DWE is FTL at its generation, but it is unknown the precise point, along the fiber, the fission of DWE occurs. The dispersion of DUV is larger than the optical frequencies for all materials, including the noble gases, therefore if the DWE is not collected at the moment it detaches from the soliton, even a short propagation through the NLO medium temporally broadens the pulse considerably. The Figure 30 shows a DUV with spectral width around 20 nm at 280 nm, which comprises pulses down to 6 fs FTL. Aiming to generate the shortest DUV pulse possible it is indispensable to compress the DUV pulse before it could be temporally characterized. We have chosen to perform SD-FROG, but other DUV temporal characterization methods are reported in the literature. Among the recently used are the difference frequency generation cross FROG (DFG-XFROG).⁶¹, which is able to characterize low intensity beams, but its experimental setup and alignment is far more complex than SD-FROG.

The DUV beam was collimated by a 1 m focus aluminum concave mirror and steered to a magnesium fluoride pair of prisms (MgF₂ - 120 – 7000 nm) compressor. As we did not know the amount of dispersion to be corrected, in our first attempt to compress the DUV pulse we have set the pair of prisms 20 cm apex-to-apex apart from each other. That distance was chosen such as we could also remove the fundamental beam by clipping it after the first prism passage, allowing just the DUV propagate along the compressor. The second passage through MgF₂ prism compressor consisted in back reflecting the beam on an aluminum plane mirror, into a slightly different downward path such as it could be removed and steered to the temporal characterization setup (Figure 32).

Figure 32 – Prism compressor. PM – Plane Mirror; P – MgF_2 Prism; S – Beam Stopper.
Source: By the author.

The SD-FROG setup is the same used previously to characterize the excitation beam (Figure 22), only exchanging the silver by aluminum mirrors. The alignment of the temporal characterization system in the DUV has shown to be challenging, considering there was no imaging system available at this wavelength. Therefore, we were not able to use the thin sapphire as NLO medium, which would be the best option considering the DUV high dispersion. Instead, we manage to observe the SD signal using a 3 mm thick sapphire that definitely temporally broadened the pulse. We first use the fundamental beam, easier to align and match the two replicas temporally, and then allowing the DUV to pass throughout the same path. Even though we were able to observe the SD signal visually and performed a temporal scan, it was not possible to record a series of spectrograms due to instabilities on the DUV intensity. A constant and significant blinking was observed on the light coming out of the SPM-HCF, and we suppose it reverberate on the temporal characteristics of the excitation pulse, by it turn was affecting the DUV generation. Plasma generation on the entrance of the SPM-HCF may be an explanation for that kind of instability, maybe due to some gas contamination on the SPM housing atmosphere. Despite the difficulty to acquire the SD measurement, we have scanned the temporal overlap between the replicas and observed a temporal width of the order of tens of fs, which is beyond the desired sub-10 fs.

For a reliable temporal characterization of the DUV pulse, the whole system, from Ti:Sapphire to the DUV generation must be stabilized, and a study of the DWE fission length must be done to extract the DWE just after is generation, because even the propagation through the DWE-HCF filling gas temporally broadened the pulse. In this sense, a pressure gradient could be created in the DWE-HCF to decrease DUV dispersion on the filling gas. A study of the distance between of the compression prisms must be performed, in order to get the shortest pulse possible. Furthermore, the whole system, after the exit of the DWE fiber should be placed inside a vacuum chamber, in order to avoid the DUV dispersion, which occurs even in air.

6 CONCLUSIONS

Through the NER measurement using hollow-core fibers, we have measured the nonlinear refraction properties of the gases that compose the atmospheric air, (namely: argon, helium, oxygen, nitrogen, along with their mixture), and characterized their dependency as a function of the temporal excitation pulse width. The NER signal is proportional to the third-order susceptibility, which by its turn is linked to $n_{2,eff}$ value differently according to the nonlinear physical mechanism. In this thesis, we have considered two origins to the nonlinear process: the instantaneous pure electronic nonresonant and the pulse width dependent molecular orientation. In noble gases, we verify that only ENR is important for its n_2 , making it independent of the temporal width of excitation beam, using this feature for its determination. We proposed an empirical model which discriminates the ENR and MO nonlinearities from the effective value, as well as a characteristic time of the MO response. The computed values for n_2 show up to be in agreement to the current accepted ones described in literature, including the NL refractive index of He, which is hardly accessed by other techniques. We also highlight the self-referenced capability of our NER measurements to determine the pulse width, avoiding the necessity of additional pulse characterization technique in order to evaluate that parameter.

For future perspective, it is necessary to study more carefully the influence of the pulse chirp on the NER measurements. So far, for the sake of simplicity, we have neglected completely. Furthermore, preliminary results in CO₂ demonstrate asymmetries, due to be a diatomic gas that deserve a deep investigation. Those studies have the potential to bring extra information about the gases regarding the ones presented in this work. Thus expanding even more the applicability of NER technique.

We successfully demonstrate an application of the nonlinear refraction of gases in the DUV generation by soliton dispersive wave emission in hollow-core fiber, with tunability varying from 150 to 350 nm. Even though, the temporal characterization could not be performed by the time due to mechanical and energy instability problems, several aspects of the experiment, ranging from dispersion management in the NIR and DUV, to a few-cycle pulse temporal characterization, going up to soliton dynamics and wave breaking phenomena have been characterized and explored. The amount of energy produced in the DUV region demonstrates the promising features of the DWE effect to generate this new source of light, which is very desirable for several applications. Its feasibility and scaling potential have already been demonstrated.

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