Theoretical and experimental investigations of ultrashort laser filamentation in gases

BÉJOT, Pierre

Abstract

La filamentation est à l'origine de transformations spatio-temporelles des impulsions laser ultrabrèves et de leur milieu de propagation. Le travail de cette thèse a été d'approfondir la compréhension de ce type de propagation. Ainsi, des simulations numériques reproduisant la propagation des filaments dans les gaz ont été développées. Expérimentalement, nous avons démontré que l'élargissement du spectre est statistiquement ordonné, permettant d'appréhender la dynamique du supercontinuum. De plus, nous avons étudié la propagation de deux filaments, la concaténation des deux canaux plasma et le fort élargissement des spectres dus aux interactions croisées des impulsions. Parallèlement, nous avons montré que les hautes intensités régnant au sein des filaments induisent une forte biréfringence, conduisant à la génération d'une lame demi-onde ultra-rapide. Enfin, nous avons étudié la propagation d'un laser de 20 J, 32 TW dans l'atmosphère, conduisant à la génération de centaines de filaments et d'un supercontinuum détectable jusque dans la stratosphère.

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Theoretical and experimental investigations of ultrashort laser filamentation in gases

THÈSE

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par

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Résumé

La filamentation est à l’origine de nombreuses transformations spatio-temporelles des impulsions laser ultrabrèves et de leur milieu de propagation. Le principal travail de cette thèse a été d’approfondir la compréhension de ce type très particulier de propagation. Ainsi, des simulations numériques, reproduisant la propagation des filaments dans les gaz, et plus particulièrement dans l’Argon, ont été entièrement développées. Expérimentalement, nous avons démontré que l’élargissement du spectre se fait de manière statistiquement ordonnée (les composantes spectrales générées sont fortement corrélées), ce qui a permis de mieux appréhender la dynamique de la génération du supercontinuum. De plus, nous avons étudié la copropagation de deux filaments et observé la concaténation des deux canaux plasma et le fort élargissement des spectres dus aux interactions croisées des impulsions. Parallèlement, nous avons montré que les hautes intensités régnant au sein des filaments induisent une forte birefringence, conduisant à la génération d’une lame demi-onde, pouvant théoriquement commuter la polarisation d’une onde en quelques centaines d’attosecondes. Enfin, nous avons étudié la propagation d’un faisceau laser de 20 J, 32 TW dans l’atmosphère, conduisant à la génération de plus de 400 filaments et d’un continuum de lumière blanche détectable jusque dans la stratosphère.

Abstract

The filamentation process induces strong spatio-temporal deformations of both the high power ultrashort pulse and the propagation medium. The main goal of this thesis was to understand thoroughly this very particular kind of propagation. Numerical simulations, reproducing the filament propagation in gases, have been entirely implemented. From the experimental point of view, we demonstrated that the spectral broadening can be explained in a corpuscular point of view and that the different spectral components are strongly correlated, which has allowed to highlight the supercontinuum generation dynamics. Moreover, we have investigated the co-propagation of two filaments and have observed the concatenation of the two plasma channels as well as the strong spectral broadening due to their crossed nonlinear interaction. We have also demonstrated that the high intensity within the filament core induces a strong birefringence, leading to the generation of an ultrafast half-wave plate, allowing to commute the polarization of a probe ultimately in a few hundreds attosecond. Finally, we have investigated the propagation of a 20 J, 32 TW laser pulse through the atmosphere, leading to the generation of more than 400 filaments and of a white-light continuum detectable up to the stratosphere.
Le jour de la soutenance et du départ du laboratoire approchant, une multitude de sentiments contradictoires m’habitent. C’est évidemment une grande joie, une grande fierté qui devraient accompagner ces derniers instants: obtenir le titre de Docteur en Physique, faire de la Science mon métier, ont été mes rêves depuis mon plus jeune âge. Mais je n’imaginais pas à quel point, ce sentiment d’accomplissement serait mêlé d’une grande tristesse : la tristesse de quitter des personnes que j’estime profondément, de quitter un Groupe d’exception.

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MERCI A TOUS !!!
Introduction

It has long been believed that intense ultrashort laser pulses are not suited for long-range propagation in air. In particular, it was believed that Kerr focusing effect would lead to a catastrophic collapse, leading in turn to the formation of a dense absorbing plasma, ruling out the propagation of such pulses. However, Braun et al. [1] reported in 1995 the first observation of self-channeling of GW laser in air along more than 20 m. This new propagation regime where the laser beam does not experience diffraction over several meters was called filamentation. Kerr-induced self-focusing and plasma defocusing were identified by the researchers as the two main mechanisms driving this particular propagation regime. The fast progress of laser technology allowed the scientific community to achieve self-channeling regimes over longer and longer distances [2, 3] and even up to 2 km [4–6]. This regime has been demonstrated both in horizontal and vertical propagation configuration and suggest even longer filamentation distances [6]. Because of the democratization of Chirped Pulse Amplifiers, filamentation rose a great interest and became a field of intense research activity [7–9] both experimentally and theoretically as it can be noticed in Fig. 1.

The main nonlinear effects experienced by ultrashort laser pulses, which are responsible for this self-guided propagation, is now well known. On one hand, the Kerr effect, due to 3rd order nonlinearity, leads the pulse to self-focus. Consequently, the intensity within the pulse core becomes higher and higher, drastically increasing the pulse intensity and, in turn, Kerr self-focusing. As the intensity reaches the ionization threshold (about $10^{14}$ W cm$^{-2}$), the generated plasma acts as a saturation mechanism, leading to the clamping of the pulse intensity. The dynamic equilibrium between these two effects induces a diffraction-free propagation over several Rayleigh lengths, which is the main feature of filamentation.

In the temporal domain, because of the combined effects of plasma ionization, chromatic dispersion, and Kerr-induced self-phase modulation, the pulse splits into several sharp peaks [10–16] and it results in the spectral domain in a tremendous broadening [11], leading to a supercontinuum spanning from the UV (250 nm) [17–22] to the IR (experimental results demonstrated a broadening up to 4 µm [9,23,24]). This spectacular property has even been used and studied in LIDAR experiments for monitoring pollutants present in the atmosphere. [25–27]. Moreover, because the supercontinuum is generated coherently, filaments have also been used to produce shorter and shorter pulse in rare gases (typically in Argon or Neon) down to the single cycle regime (2.66 fs at 800 nm). [28–39]. This extremely short duration coupled with the high intensity within the filament core has also been used to drive high harmonics generation [40] and, subsequently attosecond pulses generation [41,42].

In the spatial domain, unlike linear propagation, the dynamic equilibrium between self-
focusing and medium ionization leads to a soliton-like propagation [43] of light bullets [37, 44–46] confined within about 100 µm, and to an intensity clamping as high as about 5 \(10^{13}\) W.cm\(^{-2}\) [8, 47–49] in air or Argon. The free electron density induced by this very high intensity is about \(10^{17}\) e\(^{-}\).cm\(^{-3}\) [50–52], decreasing the local resistivity of the air to allow the triggering and guiding of high voltage discharges [53–56].

Moreover, the scalability of discharges control to outdoor experiments, should be helped because of the ability of filaments to propagate even in adverse conditions such as fog, rain, and turbulence [57–61]. These properties consequently open the way to outdoor experiments, the ultimate goal being to trigger lightnings. Even if it has not been achieved yet, recent experiments reported electric-events [62] in direct connection with the filament-induced plasma channel generated at about 100 m above the Teramobile laser source [63].

From the theoretical point of view, the fast progress in computing power has allowed to simulate more and more accurately the dynamics of such kind of nonlinear propagation [30, 43, 64–89].

Beside the rich variety of applications which have emerged from the numerous transformations experienced by high power ultrashort pulses, a lot of questions are still unanswered. In this thesis, several aspects and potential applications of filamentation have been thoroughly studied from both the theoretical and experimental points of view, for domains spanning from coherent (optimal) control in bio-molecules, to lightning triggering, from pollutants sensing, to attosecond pulse generation, and from noise reduction and ultrafast switching for remote optical communication, to phase-encrypted pulse transmission through the atmosphere.

In Chapter 1, we will describe filamentation from a theoretical and numerical point of view. In particular, the nonlinear equation driving the pulse propagation will be derived from the Maxwell’s equations. We will present the numerical scheme allowing the resolution of this
highly unstable equation. In the last part, with the help of our numerical model, we will highlight the key points of the filamentation dynamics. In particular, we will focus on the dependence of the output pulse to the different input parameters such as energy, power, focusing, chirp and gas pressure.

Chapter 2 will be dedicated to the study of pulse propagation in random or random-like media. The first section will focus on the linear propagation through a turbulent medium. In particular, we will be attached to describe the spatio-temporal distortions occurring in this fluctuating, therefore random, medium. This study is of particular interest for future remote coherent control experiments where the preservation of the phase after propagation is of prime importance. The second section will focus on the process responsible for the spectral broadening of filaments. In particular, we will focus on the physical origin of the self-phase modulation process, describing it, from a corpuscular point of view, as a series of cascading four-wave mixing.

Even if the underlying physical processes of these two propagation regimes are different, we will use in both cases, statistical tools to highlight the propagation dynamics.

Chapter 3 will focus on the nonlinear interaction between a filament and another ultrashort pulse. In the first section, we will describe how a filament can break the medium symmetry, resulting in an instantaneous birefringence. This property will be used to commute the polarization of a probe, as with an half-wave plate, but with a commutation time ultimately limited by the time duration of the driving filament. In a second section, the nonlinear interaction between two filaments will be studied. The co-propagation of two filaments will exacerbate the nonlinearities, leading to the "co-filamentation" regime where the two plasma channels and the two spectra become connected. Moreover, we will see that this particular propagation regime can lead to a cross-compression of the two pulses without any external temporal compression.

Finally, Chapter 4 will be dedicated to the scalability of the filamentation process to multi-joule, multi-Terawatt laser. We will study the propagation of such extreme laser pulses through the atmosphere. In particular, we will analyze by LIDAR measurements the white-light generated during the multi-filamentation process. This study aims at improving the knowledge of multiple filamentation in view of both white-light LIDAR and lightning control applications.
Chapter 1

Femtosecond filamentation in gases: Theoretical model and numerical simulations

This Chapter is dedicated to a theoretical description of the propagation dynamics of ultrashort and ultra intense pulses in gases. Unlike linear pulses which experience linear diffraction and dispersion only, leading to a spatio-temporal spreading after a few meters of propagation, high power laser pulses are able to sustain a very high intensity over several Rayleigh lengths without any external waveguide. A dynamic balance between two antagonist effects is responsible for such a property: self-focusing induced by Kerr effect and plasma defocusing. These two highly nonlinear effects, responsible for this self-guiding effect induce strong spatio-temporal deformations of the pulse. Unfortunately, because of the very high intensity within the pulse core, no direct experimental measurement is possible within the filament, which makes the process understanding very challenging and theoretical investigation crucial. This Chapter is composed of three main parts. In the first section, the basic equations driving the propagation will be derived from Maxwell’s theory, highlighting the two main mechanisms (Kerr effect and plasma generation) leading to the formation of the so-called ”filaments”. In the second part, we will describe the numerical implementation of the Nonlinear Schrödinger Equation (NLSE). Finally, with the help of this numerical solver, we will highlight theoretically the main spatio-temporal tendencies of the filamentation propagation dynamics. In particular, the dependence on initial conditions will be studied.

1.1 From Maxwell theory to the Nonlinear Schrödinger equation

In this section, we will focus on deriving the basic equations which drive the propagation of an ultrashort ultra intense pulse through Argon. Argon has several properties which makes its use highly relevant for a wide panel of ultrafast laser applications. First of all, Argon is a transparent and chemically stable gas (which are two necessary conditions to make it easily usable in optics
Moreover, Argon being a rare gas, all its electronic shells are full, resulting in a very high ionization potential ($U_i \simeq 15.7\, eV$) and consequently allowing transport of very high laser intensities before it ionizes. This latter property is of particular interest when dealing with laser filaments which transport intensities as high as about $10^{14}\, W \cdot cm^{-2}$. Finally, noble gas nonlinearities are purely electronic and consequently quasi-instantaneous ($\simeq 1\, fs$) compared to the usual laser pulse duration ($30 - 100\, fs$). This property will be of particular interest for further experiments (for instance, in the ultrafast Kerr gate experiment, see §3.1) where delayed effects are not desirable.

### 1.1.1 Basic equations

We consider linearly polarized real time-dependent electric field $\overrightarrow{E}$ and magnetic field $\overrightarrow{B}$. These two vectors are associated to a displacement electric field $\overrightarrow{D}$ and a magnetizing field $\overrightarrow{H}$. The relation linking $\overrightarrow{E}$ and $\overrightarrow{D}$ (respectively $\overrightarrow{B}$ and $\overrightarrow{H}$) is:

$$\overrightarrow{D} = \epsilon \overrightarrow{E} + \overrightarrow{P}$$
$$\overrightarrow{B} = \mu \overrightarrow{H}$$

(1.1)

with $\epsilon$ the medium susceptibility tensor and $\mu$ the magnetic permeability tensor, as denoted by the double underlining. These two quantities are, generally speaking, complex matrices. However, in this discussion, because Argon is an isotropic transparent dielectric medium and because only linearly polarized fields are in consideration here, $\epsilon$ and $\mu$ are assumed real scalars: $\epsilon = \epsilon_0$ and $\mu = \mu_0$. $\epsilon_0$ and $\mu_0$ follow the relation $\epsilon_0\mu_0 = 1/c^2$, where $c$ is the light velocity in vacuum.

$\overrightarrow{P}$ is the electric dipolar polarization. Both electric quadripolar polarization and material magnetization are neglected in this discussion.

In the presence of both external current density $\overrightarrow{J}$ and electric charge density $\rho$ distributions and considering all the assumptions made beforehand, Maxwell equations which govern the spatio-temporal distribution of both electric and magnetic fields are:

$$\nabla \times \overrightarrow{H} = \overrightarrow{J} + \partial_t \overrightarrow{D}$$

(1.2)

$$\nabla \cdot \overrightarrow{B} = 0$$

(1.3)

$$\nabla \times \overrightarrow{E} = -\partial_t \overrightarrow{B}$$

(1.4)

$$\nabla \cdot \overrightarrow{D} = \rho$$

(1.5)

The electric field propagation equation is found by substitution of one of the curl equations into the other, yielding:

$$\nabla \times (\nabla \times \overrightarrow{E}) = -\partial_t (\nabla \times \overrightarrow{B}) = -\mu_0 (\partial_t \overrightarrow{J} + \partial_t^2 \overrightarrow{D})$$

(1.6)

Simplifying this equation, one obtains the propagation equation of the electric field:

$$\nabla(\nabla \cdot \overrightarrow{E}) - \triangle \overrightarrow{E} + \frac{1}{c^2} \partial_t^2 \overrightarrow{E} = -\mu_0 (\partial_t \overrightarrow{J} + \partial_t^2 \overrightarrow{P})$$

(1.7)
Femtosecond filamentation in gases

For weak nonlinearities, one can show that \( \nabla (\nabla \cdot \mathbf{E}) \) behaves as \( \lambda^2 / w^2 \) where \( w \) is the transverse section of the beam. Therefore vectorial effects induced by this term can be neglected as long as the beam width is much larger than the central wavelength of the pulse. With this assumption, the propagation equation becomes:

\[
\triangle \mathbf{E} - \frac{1}{c^2} \partial_t^2 \mathbf{E} = \mu_0 (\partial_t \mathbf{J} + \partial_t^2 \mathbf{P})
\] (1.8)

In the next section, we derive the contribution of both \( \mathbf{P} \) and \( \mathbf{J} \).

### 1.1.2 Nonlinear polarization

The derivation of the polarization expression is more convenient in the reciprocal frequency space than in the temporal one. Therefore, we define \( \mathbf{P}(\mathbf{r}, \omega) \) as the Fourier transform of \( \mathbf{P}(\mathbf{r}, t) \):

\[
\mathbf{P}(\mathbf{r}, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathbf{P}(\mathbf{r}, t) e^{-i\omega t} dt
\] (1.9)

The nonlinear response of matter to intense radiation manifests itself in a nonlinear dependence of the induced polarization on the incident electric field. The nonlinearity can originate from different processes, depending on the intensity. At low and moderate intensities, the external laser field is much weaker than the static atomic Coulomb field. As a consequence, the laser field only slightly perturbs the atomic quantum states under nonresonant excitation conditions. Nonlinear interactions taking place under these conditions can be well described by a perturbative approach. Consequently, in this weak interaction regime, the nonlinear contributions constitute only a perturbation of the harmonic oscillator vision. The electric dipolar polarization is then expressed as a Taylor series of the electric field. Thus, the electric dipolar polarization is developed as:

\[
\mathbf{P}(\mathbf{r}, \omega) = \mathbf{P}_L(\mathbf{r}, \omega) + \mathbf{P}_{NL}(\mathbf{r}, \omega)
\] (1.10)

where \( \mathbf{P}_L \) (resp. \( \mathbf{P}_{NL} \)) is the linear (resp. nonlinear) medium polarization satisfying \( \mathbf{P}_{NL} \ll \mathbf{P}_L \).

In an isotropic homogeneous medium, far from any atomic resonance, the linear polarization term is proportional to the susceptibility tensor \( \chi^{(1)} \):

\[
\mathbf{P}_L(\mathbf{r}, \omega) = \epsilon_0 \chi^{(1)} \mathbf{E}(\mathbf{r}, \omega)
\] (1.11)

\( \chi^{(1)} \) is linked to the linear refractive index \( n(\omega) \) and to the wave vector \( k(\omega) \) by the relations \( n(\omega) = \sqrt{1 + \chi^{(1)}} \) and \( k(\omega) = n(\omega) \omega / c \).

The nonlinear polarization can be written as the nonlinear extension of the Taylor development of the polarization:

\[
\mathbf{P}_{NL}(\mathbf{r}, \omega) = \sum_{i \geq 2} \mathbf{P}^i(\mathbf{r}, \omega)
\] (1.12)
Since Argon is an homogeneous isotropic centrosymmetric medium, all even terms vanish. Even though several works showed that the fifth order nonlinearities can stabilize the filamentation especially in low ionization conditions [90], we will consider here only the third order nonlinearity. For electric fields all polarized along the same direction $\vec{e}$, the nonlinear polarization reads as:

$$P^{(3)}(\omega) = \epsilon_0 \iint \chi_{eff}^{(3)}(\omega; \omega_1, \omega_2, \omega_3) \prod_{j=1,2,3} (E(\omega_j)) \delta(\omega - (\omega_1 + \omega_2 + \omega_3)) d\omega_1 d\omega_2 d\omega_3 \quad (1.13)$$

where $\chi_{eff}^{(3)} = \chi^{(3)}: \vec{e} \cdot \vec{e} \cdot \vec{e}$ and $\delta$ is the Dirac distribution defined as $\delta(x) = 1$ if $x = 0$, $\delta = 0$ otherwise. This $\delta$ function takes into account the energy conservation during frequency mixing. This expression includes all four-wave mixing phenomena which respect the energy conservation principle. This expression is at the root of a link between four-wave mixing and self-phase modulation (SPM), which is currently the basic explanation of the spectral broadening during the filamentation process. This correspondence will be of particular interest in Chapter 2 to understand the underlying statistical spectral properties of filament and thus deserves to be pointed out herein. To exhibit this link, let us first simplify Eq. 1.13:

$$P^{(3)}(\omega) = \epsilon_0 \iint \chi_{eff}^{(3)}(\omega; \omega_1, \omega_2, \omega - (\omega_1 + \omega_2)) E(\omega_1) E(\omega_2) E(\omega - (\omega_1 + \omega_2)) d\omega_1 d\omega_2 \quad (1.14)$$

In this expression, one can express the time-dependent nonlinear polarization as the inverse Fourier transform of the frequency-dependent nonlinear polarization:

$$P^{(3)}(t) = \int_{-\infty}^{\infty} P^{(3)}(\omega) e^{i\omega t} d\omega \quad (1.15)$$

By shifting this integrand with ones over $\omega_1$ and $\omega_2$, the nonlinear polarization becomes:

$$P^{(3)}(t) = \epsilon_0 \int \int E(\omega_1) E(\omega_2) d\omega_1 d\omega_2 \int \chi_{eff}^{(3)}(\omega; \omega_1, \omega_2, \omega - (\omega_1 + \omega_2)) E(\omega - (\omega_1 + \omega_2)) e^{i\omega t} d\omega \quad (1.16)$$

Since the Argon response is quasi-instantaneous compared to the pulse time duration, the nonlinear temporal response can be considered as a Dirac function. $\chi^{(3)}(\omega; \omega_1, \omega_2, \omega_3)$ being the Fourier transform of the nonlinear temporal response, it can be considered as wavelength dispersion-free and simply written $\chi_{eff}^{(3)}$. Thus the time-dependent nonlinear polarization reads as:

$$P^{(3)}(t) = \epsilon_0 \chi_{eff}^{(3)} \int \int E(\omega_1) E(\omega_2) d\omega_1 d\omega_2 \int E(\omega - (\omega_1 + \omega_2)) e^{i\omega t} d\omega \quad (1.17)$$

Keeping in mind that $TF^{-1}(f(y + a)) = e^{-iat}TF^{-1}(f)$ and writing $E = \epsilon + \epsilon^*$, finally the nonlinear polarization response reads:

$$P^{(3)}(t) = \epsilon_0 \chi_{eff}^{(3)} E^3(t) = \epsilon_0 \chi_{eff}^{(3)}(\epsilon^3 + 3|\epsilon|^2\epsilon) + c.c. \quad (1.18)$$
The electric field is generally expressed as a plane wave modulated near its central frequency $\omega_0$ and wave number $k_0$. It can then be written as: $E(t) = A(t) e^{i(\omega_0 t - k_0 z)} + cc$. In this representation, if one neglects the term in $\varepsilon^3$ responsible for the frequency tripling, the nonlinear polarization can be written as:

$$P^{(3)}(t) = 6\varepsilon_0 \chi^{(3)}_{eff} |A(t)|^2 \Re e (A(t)) \cos(k_0 z - \omega_0 t)$$

This term is then responsible for SPM. It is highly efficient because it is generally considered as automatically phase matched as long as a narrow envelope is under consideration. However, when the pulse has experienced super-broadening or when dealing with ultrashort pulses, the envelope approximation cannot take place anymore and consequently, the phase-matching condition is not automatically fulfilled. The electric field has to be written as the superposition of plane waves with different wave number and pulsation: $E = \int A(\omega) e^{i(\omega t - k(\omega) z)} d\omega$. The nonlinear polarization then is expressed as:

$$P^{(3)}(t) = \varepsilon_0 \chi^{(3)}_{eff} \int \int \int (A(\omega_j) e^{i\omega_j t}) e^{-i(k(\omega_1) + k(\omega_2) + k(\omega_3)) z} d\omega_1 d\omega_2 d\omega_3 + cc$$

Hence, if a broadened pulse is under consideration, phase matching has to be taken into account. Consequently, the NLSE has to be treated more generally in the space $(k, \omega)$ to take into account all the phase matching conditions. More particularly, it turns out to be useless to add up to the fifth linear dispersion order as in [89] or even to take into account the full frequency dependence of $k(\omega)$ for the linear propagation terms without taking it into account in the nonlinear polarization term as in [37]. Neglecting the phase-matching conditions on the nonlinear polarization contribution leads to over-estimate the spectral broadening because, it is equivalent to assume that the phase matching condition is automatically fulfilled for any quadruplet $(\omega; \omega_1, \omega_2, \omega_3)$ within the supercontinuum. Moreover, the final transverse distribution of the supercontinuum is poorly estimated for the same reason. To prevent these artefact, the NLSE has to be expressed in the $(k, \omega)$ plane where $k$ is the transverse wavenumber, leading to the "X-wave" formalism [45, 91]. More particularly, the nonlinear polarization has to be expressed as $P^{(3)}(r, t) = \varepsilon_0 \chi^{(3)}_{eff} E^3(r, t)$ where $E(z, r, t) = \mathcal{H}_k(T F^{-1} \mathcal{E}(k, \omega, z))$ is the real electric field [86], $\mathcal{H}_k$ being the radial Hankel transform.

As far the frequency tripling term is concerned, linear phase matching condition is generally not satisfied along long distance. However, some works [19, 22, 88] pointed out that a nonlinear phase matching occurs during filamentation and consequently, third harmonic generation (THG) can slightly modify the propagation dynamics. More specifically, THG is responsible for an enhancement of both self-channeling and spectral broadening. Since conversion efficiency remains weak (about 1%), this phenomenon can be reasonably neglected in a first order approximation. Finally, spectral broadening of ultrashort laser generally explained by SPM corresponds alternatively to the mixing of all frequency components within the pulse spectrum. This original vision is of particular interest to understand the underlying spectral correlations which are generated during the filamentation process (see Chapter 3).
1.1.3 Ionization

Several basic laser-matter interactions can lead to electron detachment from atoms or molecules [28]. These phenomena can be sorted according to the laser intensity they require (Fig. 1.1). Exposing an atom to an intense laser field will result in a modified potential composed of the Coulomb potential and the time-dependent effective potential of the incident optical pulse. At moderate intensities, the resulting potential is close to the unperturbed Coulomb potential and an electron can be released only upon simultaneous absorption of N photons: multiphoton ionization (MPI). The MPI rate scales with the $N^{th}$ power of the intensity of the optical pulse. At sufficiently high field strengths, the Coulomb barrier narrows, allowing optical tunneling ionization to take over. At very high field strengths, the electric field amplitude reaches values sufficient to suppress the Coulomb barrier, opening the way to above-barrier ionization. However, because laser filament intensity is limited to a few tens of $TW \cdot cm^{-2}$, we will restrict our discussion to MPI. When electrons are released by ionization, the current density

$$J = q_e \rho \vec{v}_{e}$$

Figure 1.1: Typical ionization regimes depending on the strength of the applied external field. (a) In the MPI case, the external electric field is not strong enough to significantly modify the Coulomb barrier. The only possibility to free a bound electron is the simultaneous absorption of several photons so that the electron can jump over the potential barrier. (b) Tunnel ionization: when the electric field is stronger, the potential barrier is significantly modified. The barrier is fine enough to allow bound electrons to pass through the barrier by tunnel effect. (c) Above-threshold ionization: the electric field has completely suppressed the Coulomb barrier so that electrons become completely free

evolves with $q_e$ the electron charge, $\rho$ the electron density and $\vec{v}_{e}$ the electron speed which follow both the continuity equation and the fundamental principle of dynamics:

$$\partial_t \rho + \nabla \cdot (\rho \vec{v}_{e}) = \mathcal{S}$$

(1.21)

$$\partial_t \vec{v}_{e} + \vec{v}_{e} \cdot \nabla \vec{v}_{e} = \frac{q_e}{m_e} \left( \frac{\vec{E}}{c} + \frac{\vec{v}_{e} \wedge \vec{B}}{c} \right) - \nu_e \vec{v}_{e} - \frac{\mathcal{S} \vec{v}_{e}}{\rho}$$

(1.22)

where $\mathcal{S}$ represents the plasma external source, $m_e$ the electron mass and $\nu_e$ ($\tau_e = 1/\nu_e$ is about 150 fs) is the effective collision frequency of electrons. We only consider collisions with
neutral because electron-ion collisions frequency is much lower ($\tau_{ei} \simeq 10 \, \text{ps}$) in a weakly ionized plasma, while electron-electron collisions do not contribute to $\nu_e$ because the momentum of any pair of colliding electrons and associated current are conserved [92].

Combining Eq. 1.18 to 1.20, the current density dynamics can be calculated as:

$$\partial_t \vec{J} + \nu_e \vec{J} = \frac{q_e^2 \rho}{m_e} \vec{E} + \Pi$$

(1.23)

where $\Pi = \frac{q_e}{m_e} \vec{J} \wedge \vec{B} - [(\nabla \cdot \vec{J}) \frac{\vec{J}}{q_e} + (\nabla \cdot \vec{J}) \vec{J} + \vec{J} \cdot \nabla \vec{v}_e]$ represents the nonlinear ponderomotive forces. For intensities below $10^{15} \, \text{W} \cdot \text{cm}^{-2}$, this term can be neglected. Moreover, keeping terms to the lowest order in $v_e$, the plasma density dynamics is driven by the source term $S$ only. $S$ includes multiphoton ionization, avalanche ionization (by electron-neutral collisions), inverse Bremsstrahlung and electrons recombination. The plasma dynamics equation hence becomes:

$$\partial_t \rho = W(I)(\rho_{at} - \rho) + \frac{\sigma I}{U_i} - \alpha \rho^2$$

(1.24)

In this equation, $W(I)$ denotes the multiphoton ionization rate, $\sigma(\omega) = \frac{q_e^2}{m_e n_0 a_0^2 (1+\omega^2/a_0^2)}$ the inverse Bremsstrahlung cross-section, $U_i$ the ionization potential (for Argon, $U_i = 15.1 \, \text{eV}$), $\rho_{at}$ is the initial atom density and $\alpha \simeq 5 \cdot 10^{13} \, \text{m}^{-3} \text{s}^{-1}$ [68] is the recombination rate. The full derivation of the multiphoton ionization rate $W$ for any atom or ion has been derived by Keldysh [93] and alternatively by Perelomov, Popov and Terent’ev (PPT) [94].

These two theories have put forward two main regimes in direct connection with the adiabatic Keldysh parameter $\gamma$:

$$\gamma = \omega_0 \sqrt{\frac{2m_e U_i}{|q_e E_{\text{max}}|}}$$

(1.25)

where $E_{\text{max}}$ is the amplitude electric field peak. For high intensities (\gamma \ll 1\), tunnel ionization is predominant while when \gamma \gg 1, the ionization is governed by MPI. In a first approximation, the multiphoton ionization rate is directly given by:

$$W(I) = \sigma K I^K$$

(1.26)

where $K = \text{mod}(\frac{I}{I_{\text{th}}}) + 1$ is the minimal number of photons necessary to ionize an Argon atom. Obviously, multiphoton ionization processes including more than $K$ photons coexist. The full expression of the optical field ionization (OFI) rate developed by Keldysh indeed takes into account all multiphoton ionization processes. However, the ionization rate decreases very sharply with the number of photons. For instance, Fig 1.2 displays the relative contribution of the different multiphoton processes for a 800 nm laser (in the MPI approximation, $K = 11$) delivering an intensity of $10^{13} \, \text{W} \cdot \text{cm}^{-2}$. In these conditions, the ionization involving $K = 12$ photons is responsible for only 10 % of the ionization rate. The full formula for atoms or ions with quantum number $l$ and $m$ with an atomic number $Z$ is:

$$W(\omega_0, \gamma) = \omega_{a.u.} \sqrt{\frac{6}{\pi}} |C_{nl}|^2 f(l, m) \frac{U_i}{2U_H} A_m(\omega_0, \gamma)\left(\frac{2E_0}{E\sqrt{1+\gamma^2}}\right)^{2n-|m|-3/2} \exp \left(-\frac{2E_0}{3E} g(\gamma)\right)$$

(1.27)
where $A_m(\omega_0, \gamma)$ accounts for all multiphoton processes. In this equation, $n = Z(U_i/U_H)^{-1/2}$, $U_H = 13.6\, eV$ is the ionization potential of hydrogen, $\omega_{n.n.} = q_e E_H / \sqrt{2m_e U_H} \simeq 4.1 \times 10^{16}\, s^{-1}$, $E_H = q_e^5 m_e^2 / (64 \hbar^4 \pi^3 \epsilon_0^3) \simeq 501.4\, GV/m$ and $E_0 = E_H(U_i/U_H)^{3/2}$. For a given atom, the dimensionless constant $|C_{n^*,l^*}|^2$ is adapted from the hydrogen atom case by substituting the principal $n$ and orbital $l$ quantum numbers by their effective counterparts $n^* = n - \delta_l$ (where $\delta_l = n - (U_i/U_H)^{-1/2}$ is the quantum defect) and $l^* = n^* - 1$ in order to take into account the shielding induced by electrons located in the lower layers. This yields:

$$|C_{n^*,l^*}|^2 = \frac{2^{2n^*}}{n^*\Gamma(n^* + l^* + 1)\Gamma(n^* - l^*)}$$

(1.28)

where $\Gamma$ is the gamma function. The constant $f(l, m)$ reads

$$f(l, m) = \frac{(2l + 1)(l + |m|)!}{2^{|m|(|m|)!(l - |m|)!}}, \quad f(0, 0) = 1.$$  

(1.29)
The other functions involved in (1.25) read as:

\[ A_m(\omega_0, \gamma) = \frac{4\gamma^2}{\sqrt{3\pi|m|!(1 + \gamma^2)}} \cdot \sum_{K \geq \nu} e^{\exp[-\alpha(K - \nu)]} \Phi_m(\sqrt{\beta(K - \nu)}) \]

\[ \Phi_m(x) = e^{-x^2} \int_0^x (x^2 - y^2)^{|m|} e^{y^2} dy \]

\[ \beta(\gamma) = \frac{2\gamma}{\sqrt{1 + \gamma^2}} \]

\[ \alpha(\gamma) = 2 \sinh^{-1}(\gamma) - \beta(\gamma) \]

\[ g(\gamma) = \frac{3}{2\gamma}[(1 + \frac{1}{2\gamma^2}) \sinh^{-1}(\gamma) - \sqrt{1 + \gamma^2}] \]

\[ \nu_0 = \frac{U_i}{\hbar\omega_0} \]

\[ \nu = \nu_0 (1 + \frac{1}{2\gamma^2}) \]

Figure 1.3 displays the ionization rate of Argon at 800 nm and 400 nm as a function of laser intensity for both the full Keldysh theory and the MPI approximation. For 400 nm (resp. 800 nm) laser intensities up to \(10^{14}\) W.cm\(^{-2}\) (resp. \(6 \cdot 10^{13}\) W.cm\(^{-2}\)), \(\gamma \gg 1\) so that MPI approximation remains quite accurate.

![Graph showing ionization rate as a function of laser intensity](image)

**Figure 1.3:** Ionization rate as a function of laser intensity considering both 400 nm and 800 nm.

### 1.1.4 Nonlinear Schrödinger equation (NLSE)

If one considers a laser pulse linearly polarized along \(\vec{e}^x\) propagating in the \(z\) direction, with a central wavelength \(\omega_0\) and a wave vector \(\vec{k}(\omega_0) = k_0\), the associated electric field \(E(\vec{r}, t)\) can be written as:
\[ E(\mathbf{r}', t) = \sqrt{\frac{\omega_0}{2k_0}} \mathcal{E}(\mathbf{r}', t) e^{i(k_0 z - \omega_0 t)} + \text{c.c.} \]  

(1.31)

where the amplitude factor \( \sqrt{\frac{\omega_0}{2k_0}} \) allows the optical intensity to be expressed directly in \( W \cdot m^{-2} \).

Assuming that the spectral width \( \Delta \omega \) remains negligible compared to the central pulsation \( \omega_0 \), one can develop \( k(\omega) \) in a Taylor series centered around \( \omega_0 \):

\[ k(\omega) \simeq \sum_{i=0}^{N} \partial_{\omega}^i k|_{\omega_0} (\omega - \omega_0)^i \]  

(1.32)

For the electric field defined beforehand, considering the Taylor development of \( k \) up to the second order only, and assuming the slowly varying envelope approximation (SVEA) \( |\partial_{x,y,z} \mathcal{E}| \ll k_0 |\mathcal{E}| \) and \( |\partial_t \mathcal{E}| \ll \omega_0 |\mathcal{E}| \), the electric field envelope \( \mathcal{E} \) evolves according to:

\[ -i \Delta_{\perp} \mathcal{E} - i \frac{(\partial_z - i k_0)^2}{2k_0} \mathcal{E} - \left[ i \frac{k_0}{2} + k' \partial_t - i \left( \frac{k'^2}{2k_0} + \frac{k''}{k_0} \right) \partial_t^2 \right] \mathcal{E} = \frac{ik_0 n_2 T^{-2} |\mathcal{E}|^2 \mathcal{E} - i \frac{k_0}{2n_0^2 \rho_c} \rho \varepsilon - \frac{1}{2} T \sigma \rho \varepsilon}{\rho_c} \]  

(1.33)

where \( k' = \partial_\omega k|_{\omega_0} \) represents the pulse group velocity and \( k'' = \partial^2_\omega k|_{\omega_0} \) the group velocity dispersion (GVD) coefficient. \( n_2 = n_2(\omega_0) = \frac{3}{4} \chi^{(3)}(-\omega_0, \omega_0, -\omega_0, \omega_0) \varepsilon_{0} n_0^2(\omega_0) \) is the nonlinear refractive index, \( \sigma = \sigma(\omega_0) \) and \( T = I d + \frac{i}{\omega_0} \partial_t \).

Using the variable change:

\[ \tau = t - zk' \]  
\[ \xi = z \]  

(1.34)

the partial derivatives becomes:

\[ \partial_{\tau} = \partial_t \]  
\[ \partial_{\xi} = \partial_\xi - k' \partial_{\tau} \]  

(1.35)

For pulses embedding several optical cycles, \( k'/k_0 \simeq 1/\omega_0 \) and \( (I d + i \frac{k'}{k_0} \partial_{\tau}) \simeq (I d + \frac{i}{\omega_0} \partial_t) = T \). After the substitution of the variables and neglecting the second order derivative in \( \xi \) (paraxial approximation), equation (1.31) becomes:

\[ \partial_{\xi} \mathcal{E} = i \frac{\Delta_{\perp}}{2k_0} \mathcal{E} - i \frac{k''}{2} \partial_t^2 \mathcal{E} + i \frac{k_0 n_2}{n_0} |\mathcal{E}|^2 \mathcal{E} - i \frac{k_0}{2n_0^2 \rho_c} \rho \varepsilon - \frac{1}{2} T \sigma \rho \varepsilon \]  

(1.36)

Moreover, one have to take into account of the optical losses induced by ionization (multiphoton absorption (MPA)). In the MPI approximation and according equation (1.24), the electron amount \( de^- \) created in a volume \( dV = dx \cdot dy \cdot dz \) during a time \( dt \) is \( \sigma K \rho \sigma_t dV dt \). Since \( K \) photons are needed to produce a single electron, the total energy used to produce \( de^- \)
electrons in $dV$ and during $dt$ is $d^4E = -K\hbar\omega_0\sigma_K I^K \rho_{at} dxdydzdt$. Knowing that \(\frac{d^4E}{dxdydzdt} = \frac{dl}{dz}\) and that $I = \varepsilon\varepsilon^*$, one has:

$$2\frac{d\varepsilon}{dz}\varepsilon^* = -K\hbar\omega_0\sigma_K I^K \rho_{at}$$

(1.37)

Therefore, \(\frac{d\varepsilon}{dz} = -\frac{K\hbar\omega_0\sigma_K I^K \rho_{at}}{\varepsilon^*}\). Finally, if one defines $\beta^{(K)} = K\hbar\omega_0\sigma_K \rho_{at}$, one obtains the losses induced by MPA:

$$\frac{d\varepsilon}{dz} = -\frac{\beta^{(K)}}{2} |\varepsilon|^{2K-2}\varepsilon$$

(1.38)

Adding these multiphoton losses, one obtains the classical NLSE:

$$\partial_z\varepsilon = i\frac{\Delta}{2k_0} \varepsilon - i\frac{K''}{2} \partial_t^2 \varepsilon + i\frac{k_0 n_2}{n_0} |\varepsilon|^2 \varepsilon - i\frac{k_0}{2\hbar^2 n_0 \rho_e} \rho e - \frac{1}{2}(\sigma \rho e + \beta^{(K)} |\varepsilon|^{2K-2}\varepsilon)$$

(1.39)

In this equation, the nonlinear polarization induced by both ions and free electrons are neglected. The former is negligible because the ion relative density with respect to the atoms remains negligible, and because $\chi^{(3)}_{\text{ion}}/\chi^{(3)}_{\text{atom}} \approx (U_i/U_i^+)^3 \approx 20\%$ far from any absorption frequency (where $U_i$ (resp. $U_i^+$) is the ionization potential of Ar (resp. Ar$^+$)). The latter (induced by nonlinear ponderomotive forces) remains negligible if laser intensity remains much lower than $10^{15}$ $W \cdot cm^{-2}$.

### 1.2 Numerical Implementation of the 2D+1 model

The NLSE as defined in the previous section has no analytic solution and has to be solved numerically. Because the resolution of the full $3D + 1$ $(x,y,z,t)$ model needs computing power far beyond that of classical workstations, we assume a cylindrical symmetry around the propagation axis ($2D + 1$ model), drastically reducing both the numerical calculation time and the amount of memory necessary to store the data. Basically, the numerical code can be split into 7 main steps which are depicted in Fig. 1.4. After the definition of all physical constants (0), three main steps have to be implemented before the propagation loop begins. The first one is the definition of the spatio-temporal numerical grid (1). The second part of the code (2) is dedicated to the electric field definition with respect to the initial conditions (energy, focusing, chirp, pulse duration and width). The last block before the propagation loop (3) is the definition of all the linear operators (dispersion and cylindrical transverse Laplacian). These steps are described in detail below.

#### 1.2.1 Numerical grid definition

The 2D $(r,t)$ grid is completely defined by four parameters $(\Delta r, N_r, \Delta t, N_t)$ which are the spatial size, the number of spatial points, the temporal size and the number of temporal points.
respectively. The choice of these four parameters is a particularly critical issue for both the results accuracy and computational time. A numerical grid has to be defined in direct connection with the physical constants involved during the considered process. In the temporal domain, these constants are the initial pulse duration, the extremely sharpness of the growing plasma frontend (a few femtoseconds), and the final temporal distribution of the pulse which is extremely spiky. For pulse duration equal or shorter than 100 fs, the considered time domain $\Delta t$ has to be about 10 times the pulse duration. More precisely, a fixed time span of 1.2 ps has been adopted to ensure that negligible energy is located outside of the temporal grid. In order to speed up the fast Fourier transform (FFT) calculation, the number of points $N_t$ has to be a power of 2. In this case, the FFT asymptotic calculation complexity has a $O(N \log_2 N)$ behavior, while if using prime number of points, the complexity behaves as $O(N^2)$. A number of points of $N_t = 2^{13} = 8192$ is finally chosen, corresponding to 0.15 fs resolution. The choice of $\Delta t$ and $N_t$ has been made empirically: decreasing one of the two parameters leads to divergence of the code, while increasing them does not improve the final results. In the spectral domain, the temporal resolution is associated with a spectral window $\Delta \omega$ of about $3\omega_0$ at 800 nm which is sufficiently broad to account for the extreme spectral broadening occurring during filamentation. Moreover, the temporal window $\Delta t$ implies a spectral resolution $d\omega$ of about $2 \cdot 10^{-3} \omega_0$ which is accurate enough to resolve all the spectral lobes which appear during the filamentation.

In the spatial domain, the physical constants which have to be taken into consideration are: the initial beam diameter (depending on considered initial conditions), the filament size (about 100 $\mu m$ diameter) and the plasma channel narrowness ($\simeq 10 \mu m$). Because negligible energy has to lie beyond the numerical grid, we set empirically $\Delta r = 5\sigma_r$ where $\sigma_r$ is the initial $1/e^2$ intensity radius. Reducing the initial grid size leads to artificial "diffraction" on the grid edge after a few centimeters of propagation. The diffracted energy is then bounced back to the grid center and critically perturbs the calculation. A resolution of 2 $\mu m$ is sufficient to correctly resolve the plasma channel. For a pulse with an initial intensity radius $\sigma_r$ of 6 mm, the number of points necessary is therefore 15000. Using a (15000 x 8192) grid to define the electric field (which is complex and so coded on 16 bytes) requires about $15000 \times 8192 \times 16 \simeq 2 Gb$ of random access memory. Moreover other operators of the same typical size needs to be stored in the same time. The total amount of required memory would be beyond that available on typical
workstations. Moreover the calculation time would be much too long. The solution, without reducing the resolution needs, is to use an adaptive spatial grid (Fig. 1.5). The basic principle

<table>
<thead>
<tr>
<th>Initial spatial scale</th>
<th>Inadapted spatial scale</th>
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<tbody>
<tr>
<td>Propagation (focusing)</td>
<td></td>
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<tr>
<td>Intensity profile</td>
<td></td>
</tr>
<tr>
<td>∆r'</td>
<td>∆r</td>
</tr>
<tr>
<td>Useless region</td>
<td>Useless region</td>
</tr>
</tbody>
</table>

**Figure 1.5:** Principle of an adaptive resolution scheme. As soon as the size of the intensity profile is divided by 2, the electric field (real and imaginary parts independently) is oversampled by a factor 2 with a cubic spline interpolation. This method allows to significantly reduce both the calculation time and the amount of random access memory needed to store all the data.

is: as soon as the intensity profile width is divided by 2, the electric field is oversampled by a factor of 2 with a cubic spline interpolation on a region \( \Delta r' = \Delta r / 2 \), preserving the number of points and getting rid of the whole spatial region where only negligible energy remains (see Fig. 1.6). Generally speaking, a spline interpolation is used when continuity of derivatives is a particular concern which is typically our case because filamentation is a gradient-sensitive process. A spline is defined by a polynomial between each pair of data points, with its coefficients determined ”slightly” non-locally. The nonlocality is designed to guarantee global smoothness in the interpolated function up to some order of derivative. Cubic splines produce an interpolated function that is continuous through the second derivative. However this kind of interpolation works on real quantities only. Because the electric field is an a priori complex object, real and imaginary parts have to be oversampled independently. Basically, for an initial pulse radius of 6 mm, the re-scaling operation takes place three times before the filamentation onset.

### 1.2.2 Initial conditions

The initial electric field used in the numerical code is assumed to be Gaussian and is fully characterized by 5 parameters which are: pulse energy \( E \), the beam radius \( \sigma_r \) and duration \( \sigma_t \) defined at \( 1/e^2 \) intensity, the initial focusing \( f \) and an additional linear frequency chirp \( C \). These five parameters have been chosen as inputs of the code because their measure is straightforward in laboratory experiments, allowing direct comparison of the code output with experimental data. Obviously, they constitute only a rough approximation of real pulses. In the spectral domain, higher order chirp is neglected. In the spatial domain, wavefront aberrations are not taken into account even if some works show that it influences the filamentation
Figure 1.6: Example of a cubic spline oversampling. (a) (resp. (b)) corresponds to the real (resp. imaginary) component of the electric field displayed on the whole initial spatial region. (c-d): oversampling of both the real and imaginary components of the electric field in a small region around the center.

The process [95]. In fact, the only third order spatial aberration this code could take into account is the spherical one because it is the only one which have a cylindrical symmetry around the propagation axis. Moreover, when lenses are used to focus a laser beam in filamentation experiment, longitudinal and transverse chromatic aberrations occur (the focal length $f$ becoming $f(\lambda)$) which can perturb the filamentation process. However, discrepancies induced by these simplifications probably remain of second order. Moreover we will see in the following chapters that they do not prevent the numerical code to be accurate enough to qualitatively account for experimental results. Based on these assumptions, the initial electric field is in a first step defined as:

$$\varepsilon(r, t, z = 0) = \varepsilon_0 e^{-\frac{r^2}{\sigma_r^2}} e^{-i \frac{k_0}{2} r^2} e^{-\frac{t^2}{\sigma_t^2}}$$

(1.40)

with $\varepsilon_0 = \sqrt{\frac{P_{in}}{\pi \sigma_r^2}}$ and $P_{in} = \sqrt{\frac{2 E}{\pi \sigma_t}}$. Even if it can be directly introduced in the time domain, the linear chirp is added in the spectral domain for two main reasons. First, adding a spectral quadratic phase does not have to induce a change of the intensity spectrum. Secondly, experimentally chirping a pulse decreases the temporal pulse intensity. This definition of the chirp corresponds to experiments where chirp is adjusted with the help of a compressor in chirped pulse amplified (CPA) lasers. Finally, in the spectral domain, the electric field of a chirped pulse becomes:

$$\tilde{\varepsilon}(r, \omega, z = 0) \rightarrow \tilde{\varepsilon}(r, \omega, z = 0) e^{iC\omega^2}$$

(1.41)

where $\tilde{\varepsilon}$ is the Fourier transform of $\varepsilon$. With this definition, a negative (resp. positive) chirp implies that the higher (resp. lower) frequencies are emitted on the leading edge of the pulse. A positive chirp therefore corresponds to the definition of normal spectral dispersion. Figure 1.7 displays an example of initial electric field for $\sigma_r = 6 \ mm$, $\sigma_t = 30 \ fs$, $f = 1 \ m$ and $C = -500 \ fs^2$. 
1.2.3 Numerical resolution of the NLSE

Numerous schemes have been developed during the last few years [?, 11, 73, 96–100] and an extensive description of all integration schemes is beyond the scope of this section. Instead, we will briefly describe the option we have implemented. The NLSE is, computationally speaking, an initial value problem: perfectly knowing the electric field \( \varepsilon(r, t, z) \) at a distance \( z \), one wants to know what it becomes at a distance \( z + l \). We decompose the NLSE as:

\[
\partial_z \varepsilon = L_{\text{diff}}(\varepsilon) + L_{\text{disp}}(\varepsilon) + N(\varepsilon) \tag{1.42}
\]

where \( L_{\text{diff}} \) (resp. \( L_{\text{disp}} \)) represents the linear diffraction (resp. dispersion) term of equation (1.37) and \( N \) includes all nonlinear contributions. The integration code is then based on a split-step scheme in which all linear terms (diffraction and dispersion) are computed in the Fourier space (Fourier split step (FSS)) over a half step in a fully implicit scheme while the nonlinear terms are directly computed in the time domain [101]. Using a fully implicit method allows stable integration, in particular, when a tightly focused pulse propagates through its linear focus, contrary to both explicit and semi-implicit (Crank-Nicholson) schemes. Therefore, the first step reads as:

\[
\bar{\varepsilon} \rightarrow e^{ik''\omega^2dz_2/2} \cdot (Id - L_{\text{diff}}dz)^{-1} \ast \bar{\varepsilon}(r, \omega, z) \tag{1.43}
\]

where \( \bar{\varepsilon} \) represents the Fourier transform of \( \varepsilon(r, t, z) \) and \( \ast \) is the matrix product. One can notice that \( L_{\text{disp}} \propto \partial_r^2 \varepsilon \) is computed as \( e^{ik''\omega^2dz_2/2} \approx 1 + ik''\omega^2dz_2/2 \) in the Fourier space. Moreover, \( L_{\text{diff}} \) is roughly a transverse Laplacian. This operator is defined in cylindrical symmetry as:

\[
\Delta_\perp = \partial_r^2 + \frac{1}{r} \partial_r \quad (r \neq 0) \quad \tag{1.44}
\]

\[
\Delta_\perp = 2\partial_r^2 \quad (r = 0) \quad \tag{1.45}
\]

The second steps computes all the nonlinear terms as:

\[
\varepsilon(r, t, z + dz) = e^{N(\varepsilon)dz} \cdot \varepsilon \tag{1.46}
\]

This kind of computation remains valid until \( N(\varepsilon)dz \ll 1 \). However, as the pulse approaches its nonlinear focus point, the maximal intensity and related gradient increase drastically. Therefore, \( dz \) has to be kept small enough so that \( N(\varepsilon)dz \) remains small as compared to 1. In order
to keep reasonable computational duration, $dz$ is chosen adaptive because in the beginning of
the propagation, $dz$ does not need to be very small. At each propagation step, $dz$ is chosen pro-
portionally to the maximal intensity $I$ of the previous step because the nonlinear phase induced
by SPM varies as $Idz$. The initial step is fixed to 5 mm and during the filamentation process,
$dz$ is bounded to a minimal value of 50 µm.

1.3 Propagation dynamics of filamentation

1.3.1 Filamentation as a dynamic equilibrium between Kerr and plasma
effects

Observing equation (1.37), the respective effect of all the terms can be highlighted. First,
the opposite sign of the Kerr and the plasma effects shows that these two contributions have
antagonist effects on the propagation. Indeed, ionization spatially defocuses the pulse while the
Kerr effect tends to focus it. The same tendency occurs in the temporal domain. The plasma
contributes to the pulse dispersion when the Kerr effect counteracts the temporal spreading. In
conclusion, filamentation is a dynamic balance between ionization and Kerr effect. MPA and
dispersion are responsible for the arrest of the filament because these two processes tend to
decrease the pulse peak power. The first one dissipates photons to create plasma electrons and
the other one temporally spreads the pulse.

In the spatial domain, Kerr effect induces a local modification of the refractive index propor-
tionally to the local laser intensity. Hence, the modified refractive index can be roughly
written as: $n(r) = n_0 + n_2 I(r)$. Consequently, the optical path crossed by every laser portion
is a function of the local intensity. If the nonlinear index is positive (as it is in air or Argon)
and assuming, for instance, a gaussian transversal intensity distribution, the beam center slows
down compared to the pulse wings where the intensity is far lower. This results in a wave-
front curvature as with a convex lens: the beam experiences self-focusing. If the pulse peak
power exceeds the critical power ($P_{cr} = \frac{3.77\lambda^2}{8n_0n_2}$ for a Gaussian pulse), diffraction, which is the
linear saturating effect, is too weak to counteract self-focusing. If no other saturating effect
takes place, it would lead to the pulse collapse (in the paraxial approximation). Obviously, this
collapse is a pure mathematical point of view: when the intensity reaches the multiphoton ion-
ization threshold, the induced plasma acts as a defocusing lenses preventing the collapse. The
dynamic equilibrium between this two effects leads to an intensity clamping, keeping the peak
intensity quite stable over several Rayleigh lengths (Fig. 1.8(a)). Depending on initial con-
ditions, the intensity is confined in a about 100 µm structure (Fig. 1.8(b)) historically called
"filament". One can point out that, even in absence of plasma-induced saturation, the collapse
is physically never reached because of the paraxial approximation violation near the theoretical
collapse point when the pulse is too tightly focused.

1.3.2 Spatio-temporal dynamics of filamentation

During the filamentation process, the laser pulse experiences high spatio-temporal distortions.
Before ionization significantly modifies the propagation, the Kerr effect induces a self-focusing
Figure 1.8: Simulation of a 800 nm pulse propagating through 1 bar Argon (Energy: 1 mJ, duration: 30 fs non chirped and initial focusing: 1 m). (a) Dashed line: on-axis intensity during the propagation: the intensity is clamped over about 10 cm at $60 \, TW \cdot cm^{-2}$. Solid line: the plasma induced density reaches $90 \, Pe^- \cdot cm^{-3}$. (b) Quadratic radius of the pulse during the propagation. The intensity is confined over 10 cm in a 150 $\mu m$ wide filament in the temporal domain as it does in the spatial domain. Subsequently, the spectral dispersion is counteracted by Kerr effect and, just before the nonlinear focus, the temporal pulse duration drastically decreases as depicted in Fig. 1.9 (a).

As the pulse self-focuses, the strong increase of the pulse intensity enhances more and more the self-focusing effect and the fluence reaches value as high as $1.2 \, J \cdot cm^{-2}$ (Fig 1.10). When the ionization threshold is reached, the trailing edge of the pulse experiences a violent defocusing (Fig 1.9 (b) and Fig. 1.11 (b)) because of the plasma generation (Fig. 1.12 (a)). Thus the overall intensity in the spatial center of the pulse decreases and consequently the plasma density slightly decreases and is generated later (Fig 1.12 (b)). Consequently, the defocusing effect being weaker, the energy located around the filament (named energy reservoir) can focus again to create a second sub-pulse (Fig. 1.9 (b) and Fig. 1.11 (c)). The resulting global pulse is then composed of two sub-pulses. Because of strong multiphoton absorption, the first sub-pulse progressively vanishes (Fig 1.9 (b) and Fig. 1.11 (d)). In the same time, the photon bath feeds the second sub-pulse counteracting MPA and sustaining the filamentation process. When the energy reservoir is not intense enough to ensure the filamentation process, linear diffraction becomes predominant: the intensity progressively decreases (Fig 1.11 (e)). The temporal shape of the pulse is then composed of a single highly chirped peak surrounded by a quite intense pedestal. Because the spectral wings have a fixed phase relationship with the fundamental, the pulse can be post-compressed with, for instance, a grating compressor or a pulse shaper. If a perfect recompression is assumed, the resulting pulse duration amounts to $3.8 \, fs$, i.e. about 1.5 cycle (Fig. 1.13).

The strong temporal deformations which occur during the filamentation are obviously related to spectral transformations of the pulse: Kerr effect indeed induces a strong spectral broadening by self-phase modulation (SPM). More precisely, in a schematic point of view
Figure 1.9: (a) Quadratic pulse duration as a function of propagation distance. Kerr effect first self-compresses the pulse. At the nonlinear focal point, the duration has decreased by about 35% from 26 fs to 17 fs. When the ionization takes place, the pulse splits into two sub-pulses and consequently, the duration increases. In a last step, the energy bath refocuses into the center of the pulse when the two sub-pulses experience defocusing. The resulting pulse is then composed of almost a single highly chirped peak. (b) On-axis temporal intensity distribution as a function of the distance. Each distance slice is normalized by its maximum because of the high intensity range to plot.

Figure 1.10: Fluence distribution as a function of propagation distance. The white lines represent the quadratic radius of the pulse.
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Figure 1.11: Spatio-temporal dynamics of a filament in 1 bar Argon. In a first step (a), the pulse is both temporally and spatially focused by Kerr effect. (b) When the ionization threshold is reached, the trailing edge of the pulse is defocused by the induced-plasma. (c) Pulse-splitting occurs when the energy bath make the trailing edge refocuses in the center of the beam. (d) The energy reservoir refocuses near the temporal slice $t = 0$, the resulting temporal shape is then very spiky. (e) When the filamentation has stopped, the pulse is composed of almost a single highly chirped peak with a quite intense pedestal.

Figure 1.12: Spatio-temporal plasma density distribution. When the intensity reaches the ionization threshold (a), plasma becomes significative in the trailing edge of the pulse. After pulse splitting (b), the plasma is less dense near the temporal slice $t = 0$ which allows the energy reservoir to focus again.
where no spatio-temporal coupling occurs, one can write the electric field $\varepsilon(t, z)$ as:

$$\varepsilon(t, z) = \varepsilon(t, z = 0) e^{i \frac{\omega_0 z}{c}} e^{i \frac{\omega_0 z}{c} \left\langle |\varepsilon(t, z)|^2 - \frac{\rho_0}{n_0 c} \right \rangle} \varepsilon(t, z = 0) \quad (1.47)$$

Consequently, the temporal variation of the intensity induces a temporal change of the refractive index, which in turn corresponds to a variation of the instantaneous frequency and hence to a spectral broadening.

The broadening is then induced by two main effects. First, SPM which occurs in a pure Kerr medium is an intrinsically chirped phenomenon. The instantaneous frequency being linked to the temporal phase as $\omega(t) = \frac{\partial \phi}{\partial t}$, it can be written as $\Delta \omega(t)_{\text{SPM}} = -\frac{n_2 \omega_0 z}{c} \frac{\partial I}{\partial t}$. Consequently, redder (resp. bluer) frequencies are created in the leading (resp. trailing) edge of the pulse. Fig. 1.14 (a) depicts the theoretical spectrum of a 100 $fs$ pulse with a maximal intensity of $10^{13} W \cdot cm^{-2}$ propagating in Argon during 20 $cm$ compared to the initial spectrum.

The Wigner quasi-probability function defined as $W_A(\omega, \tau) = \int A(\omega + \frac{\Omega}{2}) A^*(\omega - \frac{\Omega}{2}) e^{-i \Omega \tau} d\Omega$ represents the distribution of the spectrum in the time frequency space. Fig 1.14(b) displays this distribution for the pulse associated to the broadened spectrum of Fig. 1.14(a). It clearly appears that the broadening process is intrinsically positively chirped.

The second phenomenon which induces a spectral shaping is the pulse-plasma interaction. As it happens for SPM, the instantaneous frequency shift induced by the propagation of the pulse through the plasma is approximatively given by: $\Delta \omega_{\text{plasma}}(t) = \frac{\omega_0 z}{2n_0 c} \frac{\partial \rho}{\partial t}$. Because $\frac{\partial \rho}{\partial t}$ is proportional to $|E|^{2K}$, plasma induces a blue shift of the pulse spectrum (see Fig 1.15). The combination of these two effects during the filamentation induces a strong spectral broadening, producing a supercontinuum which can span from 300 $nm$ up to 5 $\mu m$ (Fig. 1.16).

However, the spectral broadening is highly non-homogeneous within the transverse profile of the pulse because it strongly depend on the intensity. Figure 1.17 displays the spectrum of the filament along the whole beam profile 50 $cm$ after the filamentation process has stopped. It is clear that the core of the beam is much more broadened than the pulse edge, inducing an
Figure 1.14: (a) Spectral broadening induced by SPM only of a 100 fs pulse with a maximal intensity of $10^{13} \text{ W} \cdot \text{cm}^{-2}$ propagating in Argon during 20 cm (blue). The green dashed curve represents the initial spectrum. (b) Quasi-probability Wigner distribution displaying the positive chirp induced by SPM.

Figure 1.15: (a) Plasma-induced spectral blue-shifting of a 100 fs, 10 TW.cm$^{-2}$ pulse. (b) Wigner plot representing the temporal distribution of the blue-shifted spectrum. The center of the pulse, which corresponds to the highest gradient of the time-dependent plasma density (c), is highly blue-shifted.
important spatial chirp. A careful selection of the pulse core after the filament is then needed to prevent this effect for further applications.

**Figure 1.16:** Solid red: filament spectrum. Dash black: input spectrum. Filamentation induces a strong spectral broadening because of both SPM and time-dependent plasma dephasing which spans from 300 nm to 5 µm.

**Figure 1.17:** Spectral distribution along the beam profile. After filamentation, the pulse is highly chirped and concentrates the spectral broadening within its core.

### 1.3.3 Dependence on initial conditions

Since filamentation is driven by highly nonlinear processes, the propagation dynamics strongly depends on initial conditions. In this section, we will highlight global tendencies, in particular about filamentation length, plasma density and spectral broadening. The numerical simulations presented below use the initial conditions summarized in table 1.1. In each subsection, one of these parameters is modified to highlight its effect on the propagation.
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| Energy (mJ) | 1 |
| Δt_{FWHM} (fs) | 30 |
| Chirp (fs²) | 0 |
| σ_r (mm) | 6 |
| f (m) | 1 |
| Pressure (bar) | 1 |

Table 1.1: Initial conditions used for the focusing dependency

Focusing and Beam size

The propagation length before the beam collapses by self-focusing is well approximated by the semi-empirical Dawes-Marburger formula [102]:

\[ f_{\text{collapse}} = \frac{0.369k \sigma_r^2}{2\sqrt{\left(\frac{P_{in}}{P_{cr}}\right)^{1/2} - 0.858}^2} \] (1.48)

where \( k = \frac{2\pi}{\lambda_0} \) and \( \sigma_r \) is the \( 1/e^2 \) initial intensity radius. For typical energy and waist size of CPA lasers (a few mJ, < 100 fs, about 2 cm width), the typical collapse distance is about 50 m, completely out of range for laboratories experiments. Consequently, in real experiments, the pulse is generally focused to start the filamentation in laboratory length scale. The collapse location for an initial focal length \( f \) is then located at \( z_{\text{collapse}} \) which can be calculated as:

\[ z_{\text{collapse}} = \frac{f_{\text{collapse}}}{f + f_{\text{collapse}}} \] (1.49)

However, the subsequent filamentation is quite dependent on the initial focusing stage and consequently, the focal length has to be chosen carefully. Fig. 1.18 (a) depicts both the fluence and the quadratic radius (white lines) for several focal length (0.75 m, 1 m, 1.25 m, 1.5 m, 1.75 m and 2 m). Depending on the initial focusing, a second refocusing cycle can occur. More particularly, if \( f > 1.5 \) m, this refocusing takes place, enhancing the filament length but keeping the maximal fluence almost constant (about \( 1.2 \) J · cm\(^{-2}\)) and the maximal on-axis intensity to about \( 60 \) TW · cm\(^{-2}\) (Fig. 1.18 (b)). Increasing the focal length from 1 m to 2 m enhances the plasma channel length by a factor 5 but keeps the electron density more or less constant (about \( 10^{17} \) e⁻ · cm\(^{-3}\)) (Fig 1.18 (c)).

The filamentation temporal dynamics is strongly coupled to the spatial dynamics. Hence, as displayed in Fig. 1.18 (d), each focusing cycle corresponds to a minimal pulse duration. In the spectral domain, the intensity is clamped over a longer distance with increasing focal length \( f \), the spectral broadening is more efficient for longer focal length (Fig. 1.18 (e)) because \( SPM \) is a propagative effect.

Beam size

Another important parameter is the initial beam size. Indeed, according to the Dawes-Marburger formula, bigger pulses collapse further away and the number of focusing cycle is connected to
Femtosecond filamentation in gases

Figure 1.18: Influence of initial focusing. (a) Fluence as a function of the distance. When the focal length is longer than 1.5 m, a second refocusing cycle is observed. The white lines represent the beam width (quadratic radius). (b) Maximal on-axis intensity as function of the propagation distance. With the same initial conditions, intensity clamping at about $60 \cdot TW \cdot cm^{-2}$ occurs only for longer focal length. (c) Plasma density as a function of propagation distance. (d) Temporal duration as a function of the propagation distance. (e) Spectrum after filamentation. One can notice that longer focal lengths corresponds to broader spectra.
initial pulse width as can be noticed in Fig. 1.19 (a). Because focusing and beam size have quite equivalent influence on filamentation, it appears that the key parameter is the aperture number $AN = \frac{2\sigma_f}{f}$. Indeed, decreasing $AN$ improves both the filament length and the spectral broadening (Fig. 1.19 (b) and Fig. 1.19 (c)).

**Energy and Temporal duration**

Obviously, increasing the initial peak power tends to increase the filament length and the spectral broadening (Fig. 1.20 (a-b)). The key parameter concerning the pulse collapse being the peak power, an interesting point is the difference in the propagation dynamics between unchirped pulses with same peak power but with different energy and temporal duration. The spectral broadening is more efficient for shorter pulses, as depicted in Fig 1.20 (b) even if the fluence reaches a lower maximal value (Fig. 1.20 (a)). This behavior is of prime importance in the scope of white-light LIDAR application [103] (Chapter 4) where a high conversion from the fundamental into the white light continuum is needed. Hence for such applications, a shorter, less energetic source turns out to be more relevant than a long highly energetic one. This property is used in few-cycle generation experiments [31, 32, 104] where two steps are necessary to generate 2 cycles pulses. A first Argon cell is used to produce a shorter less energetic pulse,
which is in turn refocused in a second cell. A spectrum spanning more than one octave is produced by this way. In contrast, no major difference distinguishes the plasma channel density of two pulses with same peak power but different energy (Fig. 1.20 (c)).

**Figure 1.20:** Influence of Energy and pulse duration on filamentation. (a) Fluence distribution for two pulses with same peak power but different energy and duration. (b) Spectral broadening. Increasing the initial peak power induces a stronger spectral broadening. For equivalent peak power, shorter pulses are more broadened. (c) Plasma density as a function of the propagation distance for two pulses with same peak power but different energy and duration.

**Chirp**

The adjustment of both the pulse duration and the peak power during experiments with CPA lasers is generally achieved by chirping the pulse with the help of the final compression stage. However, decreasing the peak power (and increasing the pulse duration) by this method is not totally equivalent to a Fourier-transform (FT) limited laser pulse of same duration but with a narrower spectral bandwidth. Moreover, because SPM is an intrinsically chirped process, negative and chirp may have different effects even for short propagation distance (a few meters) experiments where Argon dispersion remains negligible. As depicted in Fig. 1.21, these two cases are indeed different. The filamentation length for negatively chirped pulses is longer, with a latter collapse being located before. In the spectral domain, the spectrum generated by negatively chirped pulses is significantly broader (Fig. 1.21 (b)). Such behavior cannot be described by the Dawes-Marburger formula (Eq. 1.46), which has been obtained for FT limited pulse.

For a positively chirped pulse, the redder spectral components are located in the temporal
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**Figure 1.21:** Influence of chirp on filamentation. (a) On-axis intensity as a function of the propagation distance for two pulses with same peak power but opposite chirp. (b) Spectral broadening for two pulses with same peak power but with opposite chirp. (c) Plasma density as a function of the propagation distance for two pulses with same peak power but with an opposite chirp.
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frontend of the pulse. These spectral components then experience a red shift with respect to the intrinsic chirped nature of self-phase modulation. The resulting spectrum is then more broadened and consequently, the central frequency is more depleted (Fig 1.22a). On the contrary, for negatively chirped pulses, the central frequency is reinforced because of this red shift (Fig. 1.22b). The spectral broadening as a function of the distance is depicted in Fig. 1.23 for both positively (a) and negatively chirped pulses (b).

![Figure 1.22: Spectral broadening dynamics due to self-phase modulation on positively (a) and negatively (b) chirped pulses.](image)

Gas pressure

A lot of propagation parameters are directly dependent on the pressure (see Table 1.2). Because the critical power driving the pulse collapse is inversely proportional to the pressure, one can wonder if the propagation dynamics of pulses with the same ratio $P/P_{\text{crit}}$ but in a differently pressurized gas cell is equivalent.

Fig. 1.24 (a) displays the spectral broadening for two pulses with same ratio $P/P_{\text{crit}}$ but propagating respectively in a 1 bar and 1.5 bar Argon cell. Increasing the pressure leads to an enhancement (resp. decrease) of the blue (resp. red) part of the spectrum compared to the case of a higher energy in a lower pressurized gas cell. Moreover the filament is shorter in the high pressure case as depicted in Fig. 1.24 (b) which shows the on-axis intensity as a function of the propagation distance. More particularly, the second focusing cycle does not occur, decreasing significantly the filament length. Increasing the pressure also leads to an enhancement of the electron density of the plasma channel without increasing its length (Fig. 1.24 (c)). This trend
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![Graph](image)

**Figure 1.23:** Spectral broadening as a function of the propagation distance for positively (a) and negatively chirped pulses.

<p>| | |</p>
<table>
<thead>
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<tbody>
<tr>
<td>$k''$ (fs$^2$cm$^{-1}$)</td>
<td>0.21p</td>
</tr>
<tr>
<td>$n_2$ (m$^2$W$^{-1}$)</td>
<td>3.2$p 10^{-24}$</td>
</tr>
<tr>
<td>$\beta^K$ (m$^{2K-3}W^{1-K}$)</td>
<td>3.32$p 10^{-176}$</td>
</tr>
<tr>
<td>$\sigma^K$ (s$^{-1}cm^{2K}W^{-K}$)</td>
<td>5.06$p 10^{-140}$</td>
</tr>
<tr>
<td>$\tau$ (s)</td>
<td>1.9 $10^{-13}p^{-1}$</td>
</tr>
</tbody>
</table>

**Table 1.2:** Pressure dependency of the physical constants of Argon ($p = \frac{P}{1\text{ bar}}$ is the relative pressure.)

can be well understood if one considers that during filamentation, diffraction with the help of plasma induced defocusing is in equilibrium with the self-focusing effect, which leads to:

$$\left(\frac{1.22\lambda_0}{8\pi n_0}\sigma_r^2\right)^2 + \frac{\rho(I)}{2\rho_c} = n_2I$$

(1.50)

If a peak plasma density is approximated by $\rho \simeq \sigma_K I^K \rho_{at} \sigma_t$ and assuming that a filament carries about $1 P_{cr} = \frac{3.77\lambda_0^2}{8\pi n_0 n_2^2}$ [9], the following scaling laws then predicts both the maximal intensity and associated plasma density:

$$I \simeq \left(\frac{0.76n_2\rho_c}{\sigma_K\sigma_I\rho_{at}}\right)^{1/(K-1)}$$

(1.51)

$$\rho(I) \simeq \left(\frac{(0.76n_2\rho_c)^K}{\sigma_K\sigma_I\rho_{at}}\right)^{1/(K-1)}$$

(1.52)

Then the pressure dependency of these two quantities can be evaluated: $\rho \propto p^{1-\frac{1}{K-1}}$ and $I \propto p^{-\frac{1}{K-1}}$. If one considers a 800 nm pulse ($K = 11$ in Argon), the maximal intensity...
(resp. plasma density) at 1 bar and 1.5 bar differs by 4 % (resp. 45 %), in full agreement with Fig. 1.24 (b) and Fig. 1.24 (c).

![Graphs showing](image)

**Figure 1.24:** Pressure dependency of on-axis intensity (a), spectral broadening (b) and plasma density (c).

### 1.4 Conclusion

In this Chapter, the equations driving the propagation dynamics of ultrashort ultra intense laser pulses in Argon have been derived. Hence, the dynamic balance between Kerr effect and plasma ionization self-guides the pulse without any external waveguide. The numerical integration of the NLSE enables a better understanding of the strong spatio-temporal deformations which occur during the propagation. In the spectral domain, the pulse is extraordinary broadened. Indeed, the spectrum can span on more than decade. Moreover, because of the highly nonlinear nature of the propagation, the resulting pulse structure is highly dependent on initial condition. In the last section of the Chapter, the study of these dependency enables to highlight global tendencies, which can help on the choice of initial conditions in experiments.
Chapter 2

Statistical properties of ultrashort laser pulses propagating in a transparent medium

This Chapter is dedicated to the random or chaotic propagation of ultrashort pulse since we are focusing on the propagation in turbulent medium and the modulational instability which govern filamentation. Statistics is a powerful tool for the description of such particularly random or random-like processes (i.e. complex enough to blur the underlying physics). Hence, we will use statistical tools to study and give global tendencies about two propagation regimes where the output pulse characteristics is almost unpredictable: The propagation through turbulence and the filamentation in a $\chi^{(3)}$ medium. The propagation through turbulence has been intensively studied for continuous waves or nanosecond pulses where the chromatic dispersion is not significant. Because we are interested in the propagation of ultrashort pulse through the atmosphere for spectroscopic applications, this topic is of high importance for future realistic remote experiments. Ultrashort pulses are a priori more sensitive to turbulence because their short envelope is likely to be disturbed significantly by small dephasing induced by the small temperature and pressure variations related to turbulence. In the second section of this Chapter, we will focus on the mechanism responsible for the spectral broadening of high power ultrashort pulse. In particular, we will focus on the physical origin of the self-phase modulation process, describing SPM in a corpuscular point of view as a series of cascading four-wave mixing.

2.1 Linear propagation of ultrashort laser pulse through a turbulent medium [105]

Atmospheric applications of ultrashort and ultra-intense lasers have recently emerged as a novel and very active field of research. On one hand, the use of filamentation in air as coherent white light source for Lidar (Light Detection and Ranging) remote sensing of atmospheric species has proven as an attractive method [7, 106]. On the other hand, time-resolved and coherent
control schemes have recently been used to efficiently discriminate between aerosol particles that exhibit identical spectral signatures [107, 108]. There is now a particular interest to apply control schemes, such as fluorescence depletion and coherent Raman spectroscopy [109], directly in the atmosphere, especially since the first field applications were demonstrated in the framework of the Teramobile project [7, 25]. However, transmitting an optimally shaped pulse over long distance through the atmosphere is not fully straightforward. Although optimal control can be used to control filamentation in the field [110], no demonstration of the applicability of remote quantum control of molecular species has been reported yet. Among the processes that may affect the propagation of an ultrashort laser pulse, thermal turbulence is related to be the most effective in preventing remote control techniques. In fact, contrary to dispersion and Kerr-related effects, distortions induced by turbulence cannot be avoided by a sensible choice of the pulse characteristics, or pre-compensated [63, 111] because of their random nature. Pointing variation and wavefront distortions (speckles) of nanosecond or CW laser beams induced by atmospheric turbulence have been extensively studied [112]. In this section, we investigate both experimentally and numerically how propagation through turbulence affects the properties of femtosecond laser pulses, with a particular emphasis on the temporal phase distortions encountered during the propagation of such inhomogeneous medium.

2.1.1 Experimental setup

The experimental setup is depicted in Fig.2.1. The measurements are carried out with an amplified 1 KHz Ti:Sapphire laser system delivering $2.5\ mJ, < 30\ fs$ pulses with a beam diameter of $\sim 12\ mm$ at $1/e^2$. After propagating over a distance of $d_1 = 3.8\ m$, the beam propagates through a highly turbulent region generated by the perpendicular flow of a hot air blower ($T \leq 500^\circ\ C$ with $500\ l/min$ flux, output air velocity $20\ m/s$, angular divergence $20^\circ$). The perturbation intensity is controlled by varying the distance between the blower and the beam axis. The strength of the perturbation, represented by the structure parameter of the refractive index $C_n^2$, was determined by measuring the variance in the pointing angle of a He-Ne laser collinear with the fs beam. [59] The turbulence range achievable by the experiment ($C_n^2 = 7 - 15 \times 10^{-9}\ m^{-2/3}$) represents a very strong perturbation, a few orders of magnitude higher than those typically encountered in the atmosphere. [113] After the turbulent region, the pulses propagate for an additional distance $d_2 = 3.7\ m$ before being characterized by a single shot autocorrelator (by sampling a $28\ mm^2$ portion in the center of the beam profile) or imaged by a digital CCD camera equipped with a $f = 105\ mm$ objective (spatial resolution $150\ \mu m/pixel$, 10 ms exposure time, corresponding to an integration over 10 consecutive shots). Alternatively, we characterized individual wavefront regions ($0.25\ mm^2$) by frequency-resolved optical gating (FROG) inserting in the beam path a random phase plate. This time-invariant perturbation allowed multi-shots acquisition. We measured by interferometry that the plate introduces an average phase-difference of $\sim 4\pi/3$ among two points on the wavefront separated by $2\ mm$, while an analogous estimate for a perturbation strength of $C_n^2 = 9.6 \times 10^{-10}\ m^{-2/3}$ gives a comparable value of $2\pi/3$. 
2.1.2 Theory

We have numerically simulated the propagation of a broadband ultrashort laser pulse through a turbulent medium in order to get a better insight into the mechanism which drive the pulse transformations. In particular, some pulse characteristics (for instance, the temporal phase of the pulse retrieved from Frequency Resolved Optical Gating technique) cannot be measured experimentally because the measurement technics need multishot acquisition which is forbidden due to the intrinsic fluctuating nature of the turbulence.

We consider a linear polarized electric field at $\lambda_0 = 800 \text{ nm}$. The electric field is modeled in the frequency domain as the superposition of monochromatic waves $\varepsilon(\omega)$. Each component is supposed to have the same initial wavefront curvature and radius but can have different phase:

$$\varepsilon(x, y, \omega, z = 0) = \varepsilon_0 F(x, y)G(\omega)$$  \hspace{1cm} (2.1)

with

$$F(x, y) = \exp \left( - \frac{x^2 + y^2}{2\sigma_x^2\sigma_y^2} \right) \exp \left( -ik(\omega)\frac{x^2 + y^2}{2R_{\text{curv}}^2} \right)$$  \hspace{1cm} (2.2)

and

$$G(\omega) = \exp \left( - \frac{(\omega - \omega_0)^2}{2\sigma_\omega^2} \right) \exp(-iC\omega^2)$$  \hspace{1cm} (2.3)

The propagation of each component is divided into three steps in order to match the experimental setup. First, every component propagates through the free space, supposed to be homogeneous and linear, along a distance $d_1$. Then, a Kolmogorov phase screen simulating the turbulence is applied. Finally, the components propagates again through the free space along a distance $d_2$. 

Figure 2.1: Experimental setup for the study of the temporal deformations due to the propagation through a turbulent region.
Figure 2.2: Linear filter theory of free space propagation. The electric field at a distance $z$ is deduced from the initial electric field by applying the filter $H_0(\sigma, \omega)$ in the reciprocal space.

### Pulse propagation in the free space

We supposed that the Fourier transform of each frequency component is defined in each point of the space. Therefore, the electric field $\varepsilon(\vec{r}, z = 0, \omega)$ can be associated with its angular spectral density: $\varepsilon(\vec{\sigma}, z = 0, \omega)$, where $\sigma = \left( \sigma_x = \frac{k_x}{2\pi}, \sigma_y = \frac{k_y}{2\pi} \right)$ According to the linear Maxwell theory, each monochromatic component is solution of the Helmholtz equation:

$$\Delta \varepsilon(x, y, z, \omega) + k^2(\omega)\varepsilon(x, y, z, \omega) = 0$$

(2.4)

In the scalar approximation, one can show that the spectral density for every distance $z$ can be deduced from the initial spectral density $\varepsilon(\vec{\sigma}, z = 0, \omega)$ (see Fig. 2.2) as:

$$\varepsilon(\vec{\sigma}, z, \omega) = H_0(\vec{\sigma})\varepsilon(\vec{\sigma}, z = 0, \omega)$$

(2.5)

where

$$H_0(\vec{\sigma}) = \exp \left( -ik(\omega)\sqrt{1 - 4\pi^2c^2/\omega^2(\sigma_x^2 + \sigma_y^2)} \right)$$

(2.6)

is the free space propagator. This filter is defined for $\lambda\sigma < 1$. Finally, the propagated electric field is directly calculated as the 2D inverse Fourier transform of $\varepsilon(\vec{\sigma}, z, \omega)$.

### Theory of turbulence

Atmospheric refractive index inhomogeneities perturb the wave front, inducing a distortion of both the magnitude and the phase of laser pulses propagating through the atmosphere. These inhomogeneities are associated with atmospheric turbulence such as thermal gradient, humidity fluctuations and wind shear inducing hydrodynamic instabilities. The simulation of atmospherically distorted wave fronts is an important tool for studying the effect of atmospheric turbulence on light propagation. In this section, the theory of turbulence will be briefly discussed. Even if the full theory of turbulence is beyond the scope of this section, we will present the physical basis which describe the structure of fluids in the turbulent regime. In particular, we will focus on the statistical properties of the refractive index governed by Kolmogorov statistics.
**Turbulence and Kolmogorov statistics**

In fluid dynamics, turbulent flow is a fluid regime characterized by chaotic, stochastic property changes. All the physical constants describing the propagation medium, such as humidity, pressure and temperature, fluctuate in both time and space. These fluctuations in turn generate random variations of the refractive index.

A statistical treatment has to be developed in order to account for such a stochastic nature.

The first statistical theory of turbulence, based on the notion of energy cascading and self-similarity has been proposed in 1941 by Kolmogorov [114]. This theory is based on three main assumptions about the nature of turbulence (Fig. 2.3):

- kinetic energy enters the medium on large scales $L_0$ named ”outer scale”, in the form of convection or friction on an obstacle,
- energy is transferred towards smaller scale sizes over eddy fragmentation,
- the smallest eddies spreading over the ”inner scale” $l_0$ dissipate heat and are stable.

In other words, in a turbulent flow, unsteady vortices appear on many scales and interact with each other. Turbulence causes the formation of eddies of many different length scales. The energy cascades from large-scale structures to smaller ones. This process creates smaller and smaller structures, producing a hierarchy of eddies as depicted in Fig. 2.4.

Another assumption, agreeing with experimental observations, is that the temporal fluctuations of the physical parameters follow a Gaussian random distribution. In the spatial domain, to account for the statistical distribution of the physical constants, Kolmogorov introduced the notion of ”structure function” of a stochastic variable. Basically, this function represents the mean difference between two points separated by a distance $\rho$. The structure function $D$ of a variable $V$ is then calculated as:
Figure 2.4: Example of the formation of turbulence. Smaller and smaller eddies are formed by a cascade energy from large eddies to smaller ones.

\[ D_V(\rho) = \langle |V(r) - V(r + \rho)|^2 \rangle_r \]  
(2.7)

where \( \langle \rangle_r \) corresponds to an ensemble average.

Kolmogorov theory shows, by energy conservation considerations, that the structure function of the temperature \( T \) can be written as:

\[ D_T(\rho) = C_T^2 \rho^{2/3} \]  
(2.8)

Defining the refractivity \( N \) as \( N = 10^6(n - 1) \), where \( n \) is the refractive index of the air, one can show that \( N \) depends on the pressure \( P \) and the temperature \( T \) as:

\[ N = 77.6 \left( \frac{P}{T} \right) \]  
(2.9)

The fluctuation \( \delta n \) of the refractivity then reads as:

\[ \delta n = n \left( \frac{\delta P}{P} - \frac{\delta T}{T} \right) \]  
(2.10)

In our case where the laser pulse propagates horizontally, \( \delta P \) can be reasonably neglected and the variations of the refractive index depends consequently on the temperature fluctuations only.
The structure function $D_n$ of $n$ can be derived from the temperature structure function:

$$D_n(\rho) = C_n^2 \rho^{2/3}$$  \hspace{1cm} (2.11)

where $C_n = \left( \frac{\partial n}{\partial T} \right) C_T$. The structure function of $n$ then depends on the power $2/3$ of the distance. Moreover $C_n^2$, named ”structure parameter of the refractive index”, fully characterizes the turbulence strength. Table 2.1 summarizes typical values of atmospheric turbulence.

Moreover one can show that turbulence affects the pulse pointing angle, which varies in time with a mean-square fluctuations about its average given by:

$$\sigma_\theta = 1.35 C_n \lambda^{-1/12} D^{1/2}$$  \hspace{1cm} (2.12)

where $D$ is the propagation distance. This relation is used to calibrate the turbulence induced during the experiments.

Moreover, knowing the structure function of the refractive index, one can build realistic numerical phase screen as in [115], able to account for experimental conditions. Figure 2.5(a) displays a numerical phase screen respecting the Kolmogorov distribution (Fig. 2.5(b)) for a structure constant $C_n = 10^{-5} m^{-1/3}$.

### Results and discussion

As shown in Figures 2.6(a) (experiment) and 2.6(b) (simulation), the beam profile is spatially distorted in a typical speckle pattern as a result of the propagation through turbulence. The experimental and simulated wavefront bear a strong resemblance, especially considering that the former is the superposition of 10 successive laser shots. The strongly inhomogeneous distribution of intensity, ranging from $10^{-3}$ to 2 times that of the unperturbed wavefront, results from the superposition of the multiple interferences among beam portions experiencing different phase-shifts through turbulence repeated at each wavelength in the bandwidth [116]. Beside spatial distortions, we investigated also global temporal modifications, as reported in Fig. 2.6(c). Pulse autocorrelation widths averaged over 50 independent laser shots show a clear increase with turbulence strength. The experimental data points (squares) are normalized by the duration of a pulse propagating the same distance in the unperturbed laboratory atmosphere, $\Delta \tau_{\text{unp}}$. Note that this duration does not correspond to that of a Fourier limited pulse. We derived from the simulation a comparable quantity by calculating the average of the temporal second moment of different beam portions normalized by the unperturbed value (circles). Note that, even in presence of strong perturbations, the average relative variation $\Delta \tau / \Delta \tau_{\text{unp}}$ does
Figure 2.5: (a) Phase screen simulating the local dephasing induced by the turbulence respecting the Kolmogorov distribution with a structure parameter $C_n^2 = 10^{-10}$. (b) Phase structure function for the phase screen generated is compared with the ideal phase structure function.
Figure 2.6: Experimental (a) and simulated (b) beam profile. Arrows indicate the strong (1) and a weak (2) intensity spots discussed in the text. c) Pulse duration normalized to that of a pulse propagating the same distance in absence of turbulence as a function of turbulence strength, from experiment (□) and simulation (●). Relative pulse duration as a function of pulse intensity at fixed $C_n^2 = 9.6 \times 10^{-10}$ m$^{-2/3}$ from experiment (d) and simulation (e).

not exceed 15%. The quantitative agreement between experiment and calculations over the whole range of turbulence strengths investigated, authorizes to extend the simulations to the $C_n^2 = 2 - 7 \times 10^{-9}$ m$^{-2/3}$ range, which was not accessible in the experiment.

Figure 2.6(d) and (e) compare experimental and numerical relative distortion of pulse duration as a function of the local wavefront intensity. The temporal distortions are strongly correlated with intensity, evidencing that the major deviations from the unperturbed case are concentrated in weak intensity regions. The pulse duration converges to the unperturbed one ($\Delta \tau / \Delta \tau_{\text{ unp}} = 1$) as intensity increases.

We can gather more insight by investigating locally the spectral and temporal characteristics of regions of the the beam profile with different intensities. As mentioned above, these measurements required the use of a random phase plate, which entails a much higher distortion than that generated by the hot air blower. In Fig. 2.7 we report two illustrative experimental temporal ($I(t)$) and spectral ($I(\lambda)$) profiles retrieved by multi-shots FROG measurements from
regions of different intensities: high (a and c) and low (b and d). Similarly, Fig. 2.8 displays characteristic examples of simulated $I(t)$, $I(\lambda)$, and Wigner plot of a strong (a, c, and e) and of a weak (b, d, and f) intensity region. The inspection of these traces further confirms the strong correlation between local intensity and pulse distortions. In low-intensity regions, the existence of substructures in the temporal profiles and major spectral alterations is clearly apparent from the plots. Conversely, for high intensity regions, little or no distortions are present and the traces almost perfectly overlap with those calculated for a pulse propagating without turbulence (dotted lines). The simulations allowed us to determine that in the intense spots, the effect of multiple interferences account for minor amplitude deviations in both $I(t)$ and $I(\lambda)$, not exceeding a few percent fraction of the relative intensity. These observations altogether are consistent with a scenario where interferences act similarly at all wavelengths: in a defined portion of the beam profile, the spectral components experience a similar phase-shift when passing through turbulence. Such a condition holds exclusively because of the narrow bandwidth of the femtosecond laser pulse (35 nm) as compared with the wavelength dependence of the index of refraction of air, $n_{\text{air}}$. By integrating $I(t)$ across the beam profile, we conclude that the overall pulse duration is only slightly affected by turbulence: 6% on the temporal second moment with respect to the unperturbed situation for $C_n^2 = 9.6 \times 10^{-10} \text{ m}^{-2/3}$. This $C_n^2$ value is four orders
of magnitude higher than typical atmospheric conditions. In the approximation of constant turbulence over the propagation length [117], using a standard value [113] for strong atmospheric turbulence ($C_n = 5 \times 10^{-7} \text{ m}^{-1/3}$), we can extrapolate that comparable pulse distortions should appear after kilometer-range propagation.

To conclude, the effect of atmospheric turbulence on linearly propagating femtosecond laser pulses can be simply treated as the superposition of interferences independently acting on the components of the pulse spectrum. Bright spots on the wavefront correspond to weakly perturbed regions, which essentially maintain the spectral and temporal properties of the original pulse. The degree of alteration anticipated for actual atmospheric propagation ($< 6\%$ over kilometer ranges) does not constitute an overwhelming limitation, but it is a factor to take into

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig_2_8.png}
\caption{Simulated $I(\lambda)$ (A, B), $I(t)$ (C, D), and Wigner representation (E, F) of the pulse characteristics in the high (left column) and low (right column) intensity spots on the beam profiles indicated by arrows 1 and 2 in Fig. 2.6.B.}
\end{figure}
account to design robust atmosphere remote control applications. A partial compensation of the
effects of turbulence can be achieved by adopting control strategies based on multiple-photon
interactions, because nonlinear power dependence limits higher order interactions to the re-
geons of high intensity, where the original spectral and temporal pulse features are conserved.
In analogy with what is currently done in astronomy [118], one could also use adaptive optics
to correct the wavefronts in real-time: Considering the weak spectral dependence over the pulse
bandwidth of \( n_{\text{air}} \), we suggest that this approach can compensate not only spatial, but also tem-
poral distortions of femtosecond pulses as long as the spectral bandwidth is narrow enough to
neglect the group velocity dispersion of the air.

In order to confirm these first results, further studies will explore the possibility to launch
two shaped and focused pulses (in the scope of remote pump-probe measurements) through
turbulence to simulate future pump-probe and coherent control experiments.

\subsection*{2.2 Spectral correlation as a tool for understanding the spec-
tral broadening during filamentation [119–121]}

In the previous section, the linear propagation of ultrashort pulses through a turbulent medium
has been considered and studied with statistical means. In this section, statistics will help us
to describe the highly nonlinear and chaotic propagation through a \( \chi^{(3)} \) gas of ultrashort high
power pulses. Because high nonlinearities drive the propagation of filament (ionization, Kerr
effect), the output pulse characteristics are very sensitive to the input parameters. This chaotic

ture gets the understanding of the mechanism driving the pulse propagation very complex. In
this section, we will use statistical correlations between spectral components to provide a better
insight into the filamentation process. In particular, we will focus on the SPM process. SPM
is classically treated in the temporal space. A "classical" explanation describes the spectral
broadening as an instantaneous frequency modulation. However, in this simple model, the

corpuscular point of view is eluded. Because of its \( \chi^{(3)} \) nature, SPM can be alternatively viewed
in the spectral domain as a four-photon mixing process, respecting the energy conservation
principle. In the present section, we present both experimental and theoretical results based on
the statistical properties of the broadened pulse, highlighting the corpuscular view of the SPM
process.

Considerable interest has been devoted to quantum optics and nonlinear effects in trans-
parent media. Second order nonlinearity \( \chi^{(2)} \) processes in parametric generators have been
the model of choice in this respect, where quantum photon correlations was first demon-
strated. Recent studies showed that this phenomenon also occurs for temporal solitons in optical
fibers [122, 123]. The origin of the correlations is intrinsic to the \( \chi^{(3)} \) self-phase-modulation
process [124] in the latter case.

Because the spectral broadening during filamentation is mainly due to SPM, one can expect
the same quantum phenomena to happen inside the filaments. Quantum optics of white-light
generation by SPM in the filaments and the resulting correlations in the spectrum of the white-
light continuum have not been described so far.
2.2.1 Experimental setups

A typical experimental setup is depicted in Fig. 2.9. A chirped pulse amplification CPA Ti:sapphire laser system delivered 150 fs pulses of typically 1 mJ, at 22.5 Hz repetition rate. The central wavelength of the laser was set between 805 and 817 nm depending on the alignment, with an initial bandwidth of 8 nm. The beam was focused by a spherical mirror of 5 m focal length, yielding a nonlinear focus filament onset 3 m downstream of it. This nonlinear focus was defined as origin of the propagation axis (z = 0). The filament length was 4 m.

At z = 10 m, the continuum generated by the filament was dispersed by a diffraction grating blazed on its first order with a reflectivity of 0.9 at 810 nm. Two photodiodes with a quantum efficiency of 0.93 at 810 nm selected two specific spectral channels (8 nm bandwidth each). A high efficiency of both the grating and the photodiodes is needed to minimize optical losses of the detection because quantum correlations tend to be canceled out when losses are high. The correlation coefficient between these two wavelength channels was numerically evaluated over time series of 5000 shots. Reference conditions without nonlinear propagation were obtained by using a plane mirror instead of the spherical one in order to avoid filamentation. In a second setup, the grating was replaced by a neutral achromatic diffuser on which the filament was scattered. The scattered light is analyzed by a spectrometer (Ocean Optics HR2000), providing 0.9 nm resolution between 785 and 845 nm. We recorded 5000 spectra, normalized them to unity and used them to compute the cross-correlation map across the spectrum. The cross-correlation between two wavelengths $\lambda_1$ and $\lambda_2$ was calculated as

$$C(\lambda_1, \lambda_2) = \frac{V(I_1 + I_2) - (V(I_1) + V(I_2))}{2\sqrt{V(I_1)V(I_2)}}$$

with $V(x)$ the variance of variable $x$ and $I_i$ the intensity at $\lambda_i$.

We also performed the same kind of experiments at a wavelength of 400 nm in Argon. Using a Argon-filled cell has two main advantages. First, the pressure can be adjusted and then allows a better control of the filamentation. Secondly, because Argon response is instantaneous (no Raman response), it allows to give a better insight into the origin of the correlations, separating the relative contribution of instantaneous SPM from Raman effect. A 1 mm thick BBO crystal is used for frequency doubling the fundamental wavelength. Adequate filters and dichroic mirrors reject the residual of the fundamental. The beam energy at 410 nm can be adjusted up to 1 mJ. As depicted in Fig. 2.18, the 410 nm beam was focused by a 75 cm lens inside a 1 m long cell filled with Argon. The Argon pressure was varied from 1 to 8 bar. Depending on the pressure, the filament formed close to the center of the cell and was typically a few centimeters long. As shown in Fig. 2.18(a), after about 50 cm of free propagation outside the Argon cell, the beam was scattered on the neutral target, and the light was collected with a fiber and injected into the spectrometer providing 0.6 nm resolution between 380 and 450 nm. 1000 spectra were recorded and used to compute the intensity cross-correlation maps between pairs of wavelength across the spectrum.
Figure 2.9: Experimental setup for the study of wavelength correlations.

Figure 2.10: Experimental setup for the study of wavelength correlations at 400 nm in Argon.
2.2.2 Results and discussion

Figure 2.11 displays the cross-correlation between a tunable wavelength $\lambda_1$ in the white-light continuum and one fixed wavelength at $\lambda_2 = 844 \text{ nm}$ as measured with the setup of Fig. 2.9. The central wavelength of the fundamental spectrum was $\lambda_0 = 805 \text{ nm}$ in this experiment. As expected from the quantum point of view, where two photons at the fundamental wavelength $\lambda_0$ are converted into a pair of photons at wavelengths $\lambda_1$ and $\lambda_2$, with $\frac{\lambda_0}{\lambda_1} = \frac{1}{\lambda_1} + \frac{1}{\lambda_2}$, the highest correlation coefficient $C_{\lambda_1\lambda_2} = 0.85$ is observed at the conjugate wavelength of the fixed wavelength. In contrast, the fundamental, which is depleted when the white light is generated, is anticorrelated with $\lambda_2$ ($C_{\lambda_0\lambda_2} = -0.4$). The typical number of photons measured in the experiment is $10^8$ per shot. In order to determine whether the correlations are classical or quantum, we evaluated the "Gemellity Factor" $G = \frac{F_1 + F_2}{2} - \sqrt{C_{12} F_1 F_2 + \frac{(F_1 - F_2)^2}{4}}$, where $F_1$ and $F_2$ are the Fano factors of the two wavelengths, i.e., the ratio of the signal noise at the considered wavelength over the shot noise. $G$ amounts to $10^7$, i.e. much greater than unity, for any wavelength pair within the [760-844 nm] range. Hence, although the number of detected photons is moderate and the losses in this first experiment are limited, the observed correlations are classical and represent the correlation between the instantaneous fluctuations of the different photon fluxes.

![Figure 2.11: Cross-correlation of the white-light continuum with one fixed wavelength at $\lambda_2 = 844 \text{ nm}$.](image-url)

Figure 2.12 displays the experimental map of cross-correlations of the intensity fluctuations before the onset of the filaments ((a), $z = -0.5 \text{ m}$), shortly after it ((b), $z = 0.5 \text{ m}$), and at the filament end ((c), $z = 10 \text{ m}$) as recorded with the second setup variant. Before filamentation, correlations are restricted to the region corresponding to $\lambda_1 = \lambda_2$. The regions of negative correlation are due to a slight jitter of the fundamental wavelength, as confirmed by numerical
Statistical properties of ultrashort laser pulses

**Figure 2.12:** Experimental correlation maps within the white-light spectrum at the fixed peak power of 6.5 GW (\(\approx\) twice the critical power for filamentation). (a) Before filamentation \((z = 2 \text{ m})\); (b) Filamentation onset \((z = 3.5 \text{ m})\); (c) Filament end \((z = 8 \text{ m})\).

Simulations (see below). Once filamentation occurs \((z = 0.5 \text{ m}, \text{i.e.} \text{ only } 0.5 \text{ m} \text{ of propagation within the filament, see Fig 2.12(b), SPM is initiated, and positive correlations are observed in regions corresponding to nearly conjugated wavelengths (} \frac{\lambda_2}{\lambda_0} = \frac{1}{\lambda_1} + \frac{1}{\lambda_2}). \text{ In contrast, negative correlations form a dark cross centered on the } \text{fundamental wavelength. In other words, the generation of the white-light photons requires a depletion of the same number of photons about the fundamental wavelength. This behavior is the signature for a simultaneous generation of both the “Stokes” and the “anti-Stokes” components of the continuum originating from the depletion of the fundamental wavelength of the incident laser. Further propagation of the filament (Fig. 2.12(c)) results in a more complicated structure of the correlation map, with stripes of positive and negative correlations appearing around the fundamental wavelength. These stripes are due to the typical oscillatory structure of the supercontinuum generated by } \chi^{(3)} \text{ broadening, which results from the beating of the waves generated at each wavelength in two slices of the pulse which have the same time derivative of the intensity. After the filament end, the correlation map does not evolve anymore since the laser intensity is too low to allow further } \chi^{(3)} \text{ broadening.}

The same evolution is observed when the laser power is progressively increased while the measurement is performed at a fixed position downstream of the filament \((z = 10 \text{ m, Fig. 2.13}). \text{ At low input beam power (}1.3 \text{ mJ/pulse, i.e.} \text{ 6.5 GW}, \text{ only twice the critical power for filamentation in air, Fig. 2.13(a)), the pattern typical of a single-step SPM process is observed. Increasing the laser power allows cascaded } \chi^{(3)} \text{ broadening events resulting in much more complex patterns (Fig. 2.13(b), } P = 9 \text{ GW}), \text{ and even the disappearance of the correlation between conjugated wavelengths (Fig. 2.13(c), } P = 16 \text{ GW}). \text{ These complex patterns correspond to highly structured spectra of the continuum, as displayed under the maps. These results show that intensity correlations within the spectrum of the white-light supercontinuum are generated in the course of self-guided filamentation. The absorption and (incoherent) scattering of light by the plasma which is generated within the filaments does not prevent correlations within the spectrum of the continuum.}

Similar experimental results were obtained in Argon at 410 nm. Figure 2.14 shows the experimental spectrum as a function of the argon pressure in the cell for a 0.9 GW initial peak power. As expected for a pulse above critical power (at 1 bar, the critical power, \(P_{cr} = \frac{\lambda_0^2}{2\pi n_0 n_2}\)
Statistical properties of ultrashort laser pulses

Figure 2.13: Correlation map within the white-light spectrum at a fixed position $z = 10 \text{ m}$, beyond the filament end. (a) $P = 6.5 \text{ GW}$, twice the critical power for filamentation; (b) $P = 9 \text{ GW}$; (c) $P = 16 \text{ GW}$. The panel below each correlation map displays the spectrum of the continuum at the considered power.

Figure 2.14: Experimental spectra broadened by self-phase modulation for various argon pressures. The energy was set at $150 \mu \text{J}$. The higher is the pressure, the broader would be the spectrum. At relatively high pressure, oscillations also appear in spectra.

is about $0.5 \text{ GW at } \lambda_0 = 410 \text{ nm}$, the propagation in the $1 − 7 \text{ bar}$ domain results from equilibrium between self-focusing and plasma effects. Typically, the nearly Gaussian input spectrum ($FWHM = 3 \text{ nm}$) is broadened by Kerr induced self-phase modulation, while propagating through the Argon cell. At higher pressures, the critical power decreases and therefore lowers the threshold, yielding to a larger broadening of the input pulses.

As is observed in the air, two photons from the pump at $\lambda_0$ are annihilated to produce one photon at $\lambda_1$ and its conjugated photon at $\lambda_2$, which produces correlations in the spectrum.
Figure 2.15: Experimental cross-correlation maps as a function of argon pressure for an input pulse energy of 275 µJ. Before propagation (Reference), only positive correlations are observed for fundamental wavelengths (consistent with initial overall coherent fluctuations on the incident intensity). After propagation, for low pressure (2 bar), additional positive correlations are observed for conjugated wavelengths (pairs of photons) whereas negative correlations appear between these wavelengths and input wavelengths annihilated in the $\chi^{(3)}$ process. At even higher pressure (5 bar), cascading events occur (visible through additional strips) which blur the reading of cross-correlation maps.

The occurrence of correlations essentially depends on the physical parameters involved in the propagation (input energy, argon pressure). Figure 2.15(a) shows cross-correlation maps of the white-light continuum retrieved from typically 1000 spectra for several argon pressures as well as before propagation through the cell. In this latter case ("Reference" in the figure), only positive correlations are observed. This is the signature of overall intensity fluctuations on the incident laser pulses. It indicates that fluctuations on the intensity of each wavelength are simultaneous. For propagation at low pressure (i.e. 1 bar), positive intensity correlations are observed in regions corresponding to nearly conjugated wavelength as well as in the trivial case $\lambda_1 = \lambda_2$, whereas negative correlations form a dark cross centered on $\lambda_0$. Up to 3 bar, the increase of the positive correlations area reflects further broadening of the input pulse and the creation of conjugated wavelengths. At higher pressure, cascaded events are responsible for the occurrence of new negative correlations (additional dark stripes). When the spectrum is broadened enough, one or both conjugated wavelengths are involved in a secondary event involving the third order polarization, which partially destroy the previously formed correlations.

To further assess the critical role of $\chi^{(3)}$ broadening in the generation of correlations, we performed a simple simulation of the correlation map. In this simulation, the propagation of the self-guided filament is not described in detail. Instead, only the effect of laser noise on SPM generation is considered. To simplify the calculation, we consider that the intensity within the filament is strictly clamped for a given shot, so that the SPM-generated spectrum can be calculated within one iteration. We repeated the calculation for initial intensities and wavelengths randomly fluctuating around the nominal parameters of the experiments (200 fs
Statistical properties of ultrashort laser pulses

Figure 2.16: Simulated correlation map. (a) Before filamentation ($z = 2$ m); (b) Filamentation onset ($z = 3.5$ m); (c) Filament end ($z = 8$ m).

Pulses at 815 nm, with a mean intensity of $3.3 \times 10^{16}$ W/m$^2$. The intensity was normally distributed with a standard deviation of $4.3 \times 10^{15}$ W/m$^2$, as estimated from the experimental data. Moreover, a slight jitter of 0.3 nm on the central wavelength has been considered. This fluctuation corresponds to a jitter of the central wavelength between two pixels of the spectrometer. The resulting set of 500 simulated spectra yielded a correlation map, calculated with the same procedure as for experimental data (Fig. 2.16). The calculated results are very similar to the experimental ones, showing that the dominant process is actually SPM and that the ionization does not jeopardize the correlations in spite of its higher order nonlinearity and lack of coherence (as ionization is not taken into account). Note that the large regions of positive correlation away from the fundamental wavelength stem from the fact that, away from the oscillatory spectrum around the fundamental, both wings of the continuum are generated simultaneously, all the more efficiently that the incident intensity is higher.

The numerical model described in Chapter 1 allows a further quantitative description of the spectral broadening in the course of the filamentation. Unfortunately, the calculation of the correlation coefficients with the full numerical model as calculated during experiments, which corresponds to the calculation of the final spectrum for a few hundreds of initial intensities, requires computing power far beyond that of classical workstations. Alternatively, the mechanism of cascading can be visualized when calculating the propagation cross-correlation $C_{th}$ for each wavelengths pairs:

$$C_{th}(\lambda_1, \lambda_2) = \frac{Cov(I(\lambda_1), I(\lambda_2))}{\sqrt{Cov(I(\lambda_1), I(\lambda_1))Cov(I(\lambda_2), I(\lambda_2))}}$$

(2.14)

where

$$Cov(I(\lambda_1), I(\lambda_2)) = E ([I(\lambda_1, z) - \bar{I}(\lambda_1)][I(\lambda_2, z) - \bar{I}(\lambda_2)])$$

(2.15)

$E$ being the mathematical expectation and $\bar{I}(\lambda_i)$ the average of $I(\lambda_i, z)$ over the propagation distance $L_{\text{filament}}$ calculated as:

$$\bar{I}(\lambda_i) = \frac{\int_0^{L_{\text{filament}}} I(\lambda_i, z) dz}{L_{\text{filament}}}$$

(2.16)
Figure 2.17: Correlation map for two different pressures. Solid line corresponds to couples of wavelengths $(\omega_1, \omega_2)$ which satisfy $2\omega_0 = \omega_1 + \omega_2$ (i). Left: in the low pressure regime ($P = 2$ bar), the higher correlation is reached for wavelengths couples which satisfy (i). Right: in the higher pressure regime ($P = 5$ bar), oscillations appear in the correlation map which is a characteristic of cascading processes. The higher correlations do not correspond to wavelengths couples which satisfy (i): initial correlations are partly lost.

To observe experimentally this quantity would require to record spectra at many propagation distances for an unique shot, which is fastidious. However, even if this quantity is not measurable experimentally, it indicates in what manner the spectrum is broadened during the filament propagation. The correlation maps resulting from Eq. 2.14, plotted in Fig. 2.17, for two different pressures, display two typical patterns. At low pressure, both left- and right-hand sides of the spectrum, which correspond to wavelengths induced during the propagation, are correlated. A maximum is reached for perfectly conjugated wavelengths which satisfy $2\omega_0 = \omega_1 + \omega_2$ (these wavelengths pairs are indicated by the white solid line on the map of Fig. 2.17). In contrast, the pump frequency $\omega_0$ is anticorrelated with the two sides of the spectrum. These properties confirm that, in the low pressure regime, the preponderant phenomenon is the $\chi^{(3)}$ process $2\omega_0 \rightarrow \omega_1 + \omega_2$. Above 3-4 critical power, i.e. at higher pressure (for example $P = 5$ bar in Fig. 2.17), oscillations appear in the correlation map corresponding to the generation of outer wavelengths from photons previously generated by SPM in cascading processes.

To conclude, the presence of correlations can be understood by analyzing the intrinsic nature of the SPM process. As described in Chapter 1, the nonlinear polarization describing both the third harmonic generation and SPM in the time domain corresponds alternatively to all the frequency mixing respecting the energy conservation principle in the frequency domain (see Eq. 1.13). If one considers an initial spectrum centered around a frequency $\omega_0$, the first kind of processes which occurs is $2\omega_0 \rightarrow \omega_1 + \omega_2$. These processes, converting 2 photons at $\omega_0$ into 2 photons of different frequencies, are the most efficient because the energy is initially concentrated around $\omega_0$, resulting in a depletion of the central frequency. The energy surplus is
Statistical properties of ultrashort laser pulses

preferentially transferred into the two wings of the spectrum because these modes are seeded by photons initially present in the spectrum. After this first four-wave mixing, the central frequency is depleted and the two wings are intense enough to give rise to the second step of the broadening process. The two photons at \( \omega_1 \) and \( \omega_2 \) are then transformed into two photons \( \omega_3 \) and \( \omega_4 \) (\( \omega_3 \) and \( \omega_4 \) may be \( \omega_0 \)). If the intensity is intense enough after this second step, the frequency mixing goes on and results in a broader and broader spectrum.

Hence the spectral broadening can be viewed as a central frequency modulation (SPM point of view) but it can also be viewed as a cascade of frequency mixing. This point of view highlights both the energy conservation and the spectral correlations which arise during the spectral broadening.

2.3 Spectral correlations and laser noise reduction

Negative correlations between the fluctuation of wavelengths can also be used to reduce the noise on the pulses. More precisely, the shot-to-shot noise can be represented as the variance \( \sigma^2(S) \) of the signal \( S \) recorded by the detection. Hence, if the detection integrates a spectrum over \( \Delta \lambda \), the noise can be calculated as:

\[
\sigma^2(S) = < S^2 > - < S >^2
\]

\[
= \int_{\Delta \lambda} I(\lambda_1) d\lambda_1 \int_{\Delta \lambda} I(\lambda_2) d\lambda_2 - < \int_{\Delta \lambda} I(\lambda_1) d\lambda_1 > < \int_{\Delta \lambda} I(\lambda_2) d\lambda_2 >
\]

\[
= \int_{\Delta \lambda} \int_{\Delta \lambda} I(\lambda_1) I(\lambda_2) d\lambda_1 d\lambda_2 - \int_{\Delta \lambda} I(\lambda_1) d\lambda_1 \int_{\Delta \lambda} I(\lambda_2) d\lambda_2
\]

\[
= \int_{\Delta \lambda} \int_{\Delta \lambda} \text{cov}(I(\lambda_1), I(\lambda_2)) d\lambda_1 d\lambda_2
\]

The noise therefore turns out to be the sum over the whole spectrum of the covariance of the wavelength pairs. Because the central wavelength is anti-correlated with the spectral wings (in the low pressure regime), an adequate filter rejecting as many positive covariance (correlations) as possible could significantly improve the signal to noise ratio (SNR).

Moreover, because we are interested in performing long distance optical measurements of atmospheric molecules with extremely broadband lasers, one of our direct concern is the realization of highly stable intense broadband source for more accurate remote measurements. For instance, water vapor concentration and atmospheric temperature profile measurements require a precision better than 1% to be useful for global warming models. A significant source of noise in Lidar measurements, besides atmospheric fluctuations, is laser noise. A low noise broadband laser covering several absorption bands of \( H_2O \) would therefore be an ideal source for such measurements.

Moreover, increasing interest has been recently devoted to selective excitation of biological samples [107, 125, 126] by coherent control schemes. The absorption bands of most of the relevant optically active biological molecules (tryptophan, flavins, heme molecules,...) are located...
in the UV-Visible region. However, up to now, no efficient UV-shaper has been available for this spectral region. Two-photon fluorescence induced by shaped intense ultrashort 800 nm laser pulses has been therefore applied to this scope [127, 128]. However, due to their inherently weak cross-section and nonlinear nature, two-photon based experiments need a tightly focused laser which is critical to implement for long-distance applications, even if recent advances in ultrafast lasers have shown that high-power laser light such the Teramobile [7, 63] can, under certain power and focusing conditions, create extended regions of ultra-intense illumination. The realization of UV-Visible pulse shapers [129,130] also opens the way for remote excited fluorescence with 400 nm shaped laser pulse. To achieve this goal, an intense and stable source, with a spectrum sufficiently broad to overlap a manifold of transition pathways of the target molecule, is highly desirable. The stability is an issue in particular for applications relying on closed-loop optimization schemes, such as those based on genetic algorithms [131].

Filamentation at 400 nm and 800 nm is a useful way to generate an intense broadband spectrum. However, considering the inherent high-order nonlinear nature of the filamentation process, the filament spectrum is very sensitive to the noise of the input beam, which could be a critical drawback for using filaments in connection to closed-loop optimization. In this section, we will show that the correlations which arise during the filamentation can be used to significantly reduce the shot-to-shot noise of the laser, opening the way to ultra-stable remote spectroscopic measurements.

2.3.1 Experimental setup

We used two experimental setups, similar than these used to measure the spectral correlations within the broadened spectrum. As depicted on Fig. 2.18, instead of being scattered on the neutral target, the continuum generated by the filament in argon is dispersed by a diffraction grating (order $-1$ blazed, efficiency 75% at 410 nm). It is then collimated by a cylindrical lens ($f = 4$ cm), and an iris placed in the Fourier plane of two cylindrical lenses filters out the edge frequencies of the continuum. The selected part of the beam is focused by another cylindrical lens ($f = 4$ cm) and then detected on a photomultiplier tube (PMT) connected to a boxcar amplifier. Another PMT, located before the argon filled cell, is dedicated to measure simultaneously the laser input fluctuations. Acquisitions of about 2000 laser shots is performed for each pressure and/or each position and size of the iris in order to retrieve histograms of the signal prior and after nonlinear propagation in the argon filled cell. Parameters maximizing the Signal-to-Noise Ratio is determined from these histograms. The same kind of experiments have also been performed in the air at 800 nm.

2.3.2 Results and discussion

Laser noise reduction in Argon

Fig. 2.19 shows histograms of the signal intensity before and after propagation through 2 bar Argon, both with and without spectral filtering over the [406-414nm] range. As compared to the incoming beam, filamentation yields a SNR increase of about 7 dB for adequate spectral
Figure 2.18: Noise reduction setup: After their propagation, laser pulses are dispersed onto a grating before entering a spectral filtering setup composed of an adjustable iris in the Fourier plane of two cylindrical lenses. The filtered broaden spectrum (collected on \( PMT_2 \)) is then compared to a sample of the input spectrum (\( PMT_1 \)) in order to observe noise reduction as a function of the argon pressure in the cell.
Figure 2.19: Histograms depicting the statistics over 2000 shots of the pulse intensities fluctuations before and after propagation in a cell filled with 2 bar of argon gas. The average energy was set at 275 µJ. (a) Without spectral filtering. No noise reduction is observed. (b) Same experiment made with a [406nm-414nm] filter. An increase of 7 dB on the SNR is clearly visible.

filtering. This noise reduction disappears if spectral filtering is applied only to one side of the spectrum because SPM rejects the fluctuations to both edges of the spectrum. Moreover, a careful study of the correlation map shows that the maximal noise reduction achievable in these condition is about 9 dB with a spectral filtering spanning over [401-412nm].

Noise compression can also be visualized by plotting the intensities of the spectrally filtered continuum versus the intensities on the channel before filamentation. In fig. 2.20, such plots are given for several Argon pressures. For pressures lower than 2.5 bar, the output intensity depends linearly on the input intensity \( I_{\text{output}} \simeq 0.9 I_{\text{input}} \).

At these pressure, the broadening does not take place, so that no spectral component is filtered out. Above 2.5 bar, the evolution of the filtered output signal is no longer proportional to the input. Instead, a plateau appears for a range of input intensities (between 1.5 and 2.5 arbitrary units in Fig. 2.20). Three intensity regimes can then be defined, labeled from I to III. In regime I, the incident intensity is too low and consequently SPM generation is not efficient enough to induce noise reduction. Regime II corresponds to the range of input intensities for which noise reduction is optimal. Although the input intensity increases by a factor 2, the spatially filtered output intensity remains stable. The signal-to-noise ratio is increased up by a factor to 5 in this interval. Pairs of incident photons at the fundamental wavelength are efficiently converted in pairs of conjugated photons by FWM which results in a reduction of the noise on the incident spectrum. In region III, cascading processes take place which result in a further increase of the noise.

Laser noise reduction in the air
Figure 2.20: Dependence of filtered intensities after propagation on the input intensities for several argon pressures. The average energy was set at 120 µJ. At low pressure (2 bar), an increase of the input intensity yields to a similar linear increase in the output intensity. At higher pressure, the behavior can be divided in 3 intensity regions. In the low intensity case (I), the power is too low to create many pairs of conjugated photon and induce a significant noise reduction on the input wavelengths. The intermediate input intensities case (II) is the region where noise reduction is the highest. For a fluctuation $\Delta S_1$ on the input intensity, a reduction of about 5 is observed on $\Delta S_2$. In region III (high intensity cases), noise reduction is lost because cascaded events start to add wavelengths to the broadening, and generate much more complex correlation maps.
Analogous results have been obtained in the air with 800 \( nm \) filaments. However, the SNR increase after spectral filtering is not as high as those obtained in more controlled conditions in Argon. An increase of 1.2 \( dB \) has been obtained with a [785-820nm] spectral filter. In order to estimate the highest level of compression that would be achievable using filaments, we sought for an optimal filter, restricting ourselves to square bandpass filters within the [750-880nm] spectral range, and considering experimental data with 6.5 \( GW \) peak power, corresponding to Fig. 2.12. The spectral range yielding maximal noise reduction is [809-819 nm], \( i.e. \) centered on the incident laser wavelength (815 nm). This range is independent of the propagation distance within the filament. The noise reduction over this optimal spectral range increases from 0.69 \( dB \) shortly after the filament onset, to 3.6 \( dB \) at the filament end.

2.4 Conclusion

In this Chapter, we have used statistical tools to provide a better insight into the physical mechanisms which drive the ultrashort pulse propagation in random or chaotic situations. First, the effect of atmospheric turbulence on ultrashort pulses has been studied. We demonstrated both experimentally and numerically that even if the propagation through turbulence induces strong spatial wavefront deformations, the temporal shape is preserved within the most intense spots of the intensity pattern. Hence, the turbulence should not prevent multi-photon experiments based on coherent control schemes where the preservation of the temporal phase is critical. Moreover, the dispersion of the refractive index of the air is low, so that it leads us to expect that a correction of the spatial wavefront should simultaneously correct the temporal deformations experienced by the lower intense spot.

In the second part of this Chapter, we have showed that correlations are generated during the spectral broadening. From the fundamental point of view, these correlations allow a better understanding of the SPM process: SPM can be alternatively seen as a cascade of four-wave mixing events where all the processes \( \omega_1 + \omega_2 \rightarrow \omega_3 + \omega_4 \) progressively occur during the filamentation.

Moreover, by an adequate spectral filtering, these correlations can be used to significantly reduce the shot-to-shot noise. We experimentally demonstrated a noise reduction of 7 \( dB \). This noise reduction could be applied when an ultra-stable broadband intense laser pulse is required \( e.g. \) for remote closed-loop spectroscopic applications.
Chapter 3

Nonlinear interaction between filaments and ultrashort pulses

Because of the very high intensity within the filament core, a co-propagating probe beam can be significantly affected. Up to now, studies of filament-induced changes in the refractive index of air have only considered long-lasting effects (nanoseconds to microseconds) of the plasma left behind the pulse [132, 133]. In this Chapter, two main instantaneous interactions will be highlighted. In the first section, filament induced birefringence will be discussed. Self-induced birefringence has been observed to generate refractive index changes $\Delta n$ in the $10^{-5}$ range for ultrashort lasers focused into gases [134]. The resulting phase shift remains, however, marginal as diffraction restricts the effect to a Rayleigh length of about 100 $\mu$m around the beam waist position. We will see that the unique ability of a filament to carry a very high intensity during several Rayleigh lengths can generate an ultra-fast half-wave plate, opening the way to long distance logical gating. In the second part of the Chapter, the co-filamentation of 400 nm and 800 nm ultrashort pulses will be studied. In particular, we will describe the propagation dynamics which drives the co-propagation of such ultra-intense pulses. Such an interaction could be useful for applications such as few-cycle pulse generation for attosecond science because of the tremendous broadening induced by the interaction of the two filaments coupled to a cross-compression effect. It may also generate a very long plasma channel suitable for lightning control experiments.

3.1 Nonlinear birefringence induced by filamentation: towards ultrafast logical gating in gases [135]

An important component of high-speed optical communication networks is an all-optical switch, where an incoming “switching” beam redirects other beams through interaction in a nonlinear optical material. Among various optical schemes such as interferometers, optical polarization switches have received much interests and been demonstrated for all-optical signal processing due to the promising potential of performance of high-nonlinearity in compact scheme. However, the current available technologies use solid-state media where the nonlinear response
Nonlinear interaction between filaments and ultrashort pulses

is not instantaneous. This delayed response results in a commuting time intrinsically limited by the optical response duration ($\simeq 1\,\text{ps}$). Unlike most media, Argon has an instantaneous electronic nonlinear response only, even if its nonlinearity is very weak. Hence, Argon could potentially be a good candidate for "ultimate" optical switching. Moreover, because filament can carry very high intensity over a long distance, its use as a driving pulse can compensate the very weak nonlinearity of Argon. The results presented in this section open the way to optical switching with a commuting time as short as the electronic nonlinear response of Argon ($\simeq 1\,\text{fs}$).

3.1.1 Experimental setup

A sketch of the experiment is displayed in Fig. 3.1. A slightly chirped driving pulse (800 nm, 1 mJ, 100 fs) and a probe (400 nm, 1 µJ, 100 fs), both linearly polarized and slightly focused ($f = 1\,\text{m}$), propagate collinearly in an Argon-filled cell of 2 m length. While the driving pulse generates a single filament, the probe pulse propagates linearly in absence of the driving pulse. The relative delay between the two input pulses is adjusted by a delay line, while a zero-order half-wave plate is used to set the input polarization of the driving pulse, taken as the reference (x axis, $\theta = 0^\circ$). At the output of the cell, the intensity of a small portion of the probe beam is selected by a pinhole (1 mm diameter) and measured through a polarizer (Glan cube). The signal recorded as a function of the polarizer orientation, bears both the ellipticity and the polarization axis of the output probe pulse. A polarization state can be expressed in the Jones formalism as

![Figure 3.1](image_url)

**Figure 3.1:** Experimental setup for induced birefringence measurements. The probe beam co-propagates with the filament in the Argon cell, with an adjustable linear polarization. At the cell output, a Glan cube polarizer is used as an analyzer. A spatial selection is made afterwards to prevent artefacts due to the inhomogeneous probe polarization after the interaction with the filament.
a vector $\vec{J} = \begin{pmatrix} \cos \alpha \\
 \sin \alpha e^{i \varphi} \end{pmatrix}$, $\alpha$ and $\varphi$ characterizing the polarization state of the pulse [136]. The output analyzer with its main axis at an angle $\theta$ relative to the linear polarization of the filament is expressed as a $2 \times 2$ matrix $A = \begin{pmatrix} \cos^2 \theta & \sin \theta \cos \theta \\
 \sin \theta \cos \theta & \sin^2 \theta \end{pmatrix}$ which acts on the Jones vector $\vec{J}$. The polarization Jones vector describing the pulse polarization state after it has propagated through the polarizer is therefore: $\vec{J}_{\text{polarized}} = \begin{pmatrix} \cos^2 \theta \cos \alpha + \sin \theta \cos \theta \sin \alpha e^{i \varphi} \\
 \cos \theta \sin \theta \cos \alpha + \sin^2 \theta \sin \alpha e^{i \varphi} \end{pmatrix}$. $\alpha$ and $\varphi$ can consequently be retrieved by fitting the experimental polar plot.

### 3.1.2 Experimental results

When the filament co-propagates with a probe, with adequate relative polarization angle and delay between the two pulses, a probe signal is recorded orthogonally to its initial polarization: This is a clear signature of filament-induced birefringence. The probe spectrum is recorded as a function of the analyzer angle. Figure 3.2 displays polar plots obtained for several pressures (colored squares) for an initial polarization angle of 55° between the drive and the probe beam. It depicts the probe intensity as a function of the analyzer angle.

A fit of the angular pattern of the output polarization (Fig. 3.2) yields a contrast ratio of 67.7% at 1 bar, 39.5% at 2 bar and 98.4% at 3 bar, as well as the orientation of their ellipticity main axis, which amounts to $-51 \pm 3^\circ$, $71 \pm 5^\circ$ and $57 \pm 2^\circ$ respectively.

![Figure 3.2: Polar plot of the probe intensity as a function of the analyzer angle for several Argon pressure. 0° corresponds to the driving pulse polarization axis.](image)

The Jones parameters $\alpha$ and $\varphi$ fitting the experimental results are summarized in Tab. 3.1. The solid lines in Fig. 3.2 correspond to the associated theoretical polar plot fitting the experimental results.

The interaction with the filament is described by a dephasing operator $F$ expressed in Jones formalism as: $\begin{pmatrix} e^{-i \Phi / 2} & 0 \\
 0 & e^{i \Phi / 2} \end{pmatrix}$. $\Phi$ being the induced probe dephasing. The probe being polarized linearly with an initial angle $\gamma$ relatively to the filament polarization, it is described as: $\begin{pmatrix} \cos(\gamma) \\
 \sin(\gamma) \end{pmatrix}$. 
Nonlinear interaction between filaments and ultrashort pulses

<table>
<thead>
<tr>
<th>Pressure (bar)</th>
<th>$\varphi$ (rad)</th>
<th>$\alpha$ (rad)</th>
<th>Dephasing ($\lambda$ unit)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Input</td>
<td>0</td>
<td>-0.96</td>
<td>0.11</td>
</tr>
<tr>
<td>1</td>
<td>1.43</td>
<td>-0.42</td>
<td>0.11</td>
</tr>
<tr>
<td>2</td>
<td>-1.18</td>
<td>3.99</td>
<td>0.29</td>
</tr>
<tr>
<td>3</td>
<td>0.253</td>
<td>4.31</td>
<td>0.48</td>
</tr>
</tbody>
</table>

Table 3.1: Jones coefficients (second and third columns) retrieved by fitting the experimental polar plots for several pressures. The fourth column shows the dephasing induced between the two polarization axis in $\lambda$ unit.

Knowing both the polarization state of the probe at the output and at the input of the cell, $\Phi$ is retrieved, giving the birefringence due to the transient cross-Kerr effect, as given in the fourth column of Table 3.1.

Figure 3.3: Experimental (circles) and simulated (triangular) pressure dependence of the filament-induced dephasing.

Figure 3.3 depicts the dephasing between the two polarization axes induced by the filament, as a function of the pressure. It shows a clear linear dependence. With the initial conditions described above, a remarkable situation occurs at $P = 3$ bar. After the interaction, the probe beam polarization remains highly linear with a contrast ratio as high as 98.4%, but rotated to $57 \pm 2^\circ$, symmetrical to the initial one with regard to that of the filamenting pump beam. This probe polarization flipping is the same as would be expected with a half-wave plate inserted in the probe beam path with its neutral axis at $0^\circ$. More precisely, the observed filament-induced birefringence corresponds to a probe/2.1 ”waveplate”, i.e. the difference in the optical paths between the parallel and the perpendicular components of the probe beam amounts 1/2.1 optical cycle. Such a remarkably large dephasing therefore provides a way to tilt the linear polarization of an ultrashort laser pulse by a controlled amount: The angle of rotation of the probe polarization is twice the angle that is initially set between the input probe pulse polarization and the driving beam polarization, which undergoes filamentation, as shown in Fig. 3.4. Notice, in par-
Figure 3.4: (a) Polar plot of the filament intensity as a function of the analyzer angle. The filament is linearly polarized at 0°. (b) Probe polarization rotation at 3 bar. After the interaction with the filament, the probe is flipped symmetrically with respect to the filament polarization. (c) Polarization rotation as a function of the relative angle between the probe and the filament. The filament acts as a half-wave plate.

ticular, that choosing a 45° angle between the polarizations of the driving and probe beams flips the probe polarization by 90° at the cell exit. Setting the polarizer perpendicular to the initial probe polarization allows then to switch the probe beam intensity on and off. Hence an angle of 45° between the filament and the probe polarizations allows the realization of an ultrafast Kerr-gate able to switch the probe polarization. Since the nonlinear response of Argon is electronic, hence instantaneous, the switching time is ultimately limited by the duration of the filamenting pulse. In particular, if the probe is longer than the driving filament duration, the ultrafast half-wave plate can be used to selectively switch a short temporal slice within the probe pulse. Hence, fig. 3.5 displays the spectrum recorded with the analyzer set orthogonal to the initial probe polarization as a function of the time delay. Depending on the relative delay between the two pulses, the recorded spectrum is slightly chirped. Because the 400 nm pulse is initially positively chirped, this constitutes an indirect piece of evidence of the time gating property of the filament: The filament selects only the temporal slice of the probe co-propagating with it.

3.1.3 Discussion

Two pulses $E_1$ and $E_2$ which co-propagates in a $\chi^{(3)}$ medium are coupled by cross-phase modulation (XPM). Hence the high intensity carried by a filament induces a transient refractive index change which is experienced by any electric field co-propagating with it. Moreover, the nonlinear modification of the medium induced by a filament, which is linearly polarized is a
**Figure 3.5:** Frequency resolved polarization of the probe beam gated by the interaction with the filament. The probe spectrum is recorded with the analyzer orthogonal to the initial probe polarization as a function of the relative delay between the filament and the probe. A negative delay corresponds to the case where the filament is launched before the probe. The negative slope indicates a positive chirp of the gated slice of the probe. Because the probe is initially positively chirped, this constitutes an indirect piece of evidence of the temporal gating property of the filament.

Priori non isotropic and then can break the symmetry in the optical response of the otherwise isotropic Argon gas. This anisotropy stems from the difference between the components of the $\chi^{(3)}$ tensor: bounded electrons oscillate along with the extremely strong filament field which stretches the formerly spherical electron cloud into an ellipsoid elongated along the polarization direction of the filament. This elongation typically increases the refractive index along the direction of the filament polarization and decreases it for the orthogonal direction.

In general, the third-order susceptibility is a fourth-rank tensor with 81 elements. In an isotropic medium, such as Argon, the symmetries imply that only three elements are independent of one another, and the third-order susceptibility can be written in terms of them as:

$$\chi_{ijkl}^{(3)} = \chi_{xxx}^3 \delta_{ij} \delta_{kl} + \chi_{yyyy}^3 \delta_{ik} \delta_{jl} + \chi_{xyxy}^3 \delta_{il} \delta_{jk}$$

(3.1)

where $\delta_{ij}$ is the Kronecker delta function.

Hence if an intense pump pulse $E_R$ polarized along $\vec{e}_R = \hat{e}_x$ and a weak probe beam $E_B$ linearly polarized along $\vec{e}_B$ co-propagate, only one term remains in the expression of the nonlinear polarization along the two main axis $\vec{e}_x$ and $\vec{e}_y$. They are written respectively:

$$P_{XPM,x}^B = \frac{3e_0}{2} \chi_{xxx}^3 |E_R|^2 E_{B,x}$$

(3.2)

$$P_{XPM,y}^B = \frac{3e_0}{2} \chi_{xyxy}^3 |E_R|^2 E_{B,y}$$

(3.3)
All the other non-linear Kerr polarization components are negligible here, as the ratio of the probe and driving beam energies is $10^{-3}$. As the dominant contribution to the $\chi^{(3)}$ tensor is of electronic origin and considering that filament and probe frequencies are far from any resonant transition, one can consider that $\chi^{(3)}_{xxxx} = 3\chi^{(3)}_{yyxx}$ [137]. As a consequence, the filament induced birefringence is:

$$\Delta n = \frac{1}{2n_0} \left( \frac{3}{2} Re(\chi^{3}_{xxxx} - \chi^{3}_{yxx}) \right) |E_x|^2 = n_2^{XPM} I_{filament}$$

(3.5)

where $n_2^{XPM} = \frac{4}{3} Re(\chi^{3}_{xxxx})/(n_0^2\epsilon_0c) = \frac{4}{3} n_2^{SPM}$ represents the XPM nonlinear refractive index. As is usual in filamentation models, the third-order susceptibility of the plasma (ions and electrons) is not taken into account, because (i) electrons only have a significant contribution in the relativistic regime [138] and (ii) the small relative abundance of ions relative to neutral molecules (typically $10^{-4}$ [49]), which is not reached in this experiment and the fact that their third-order susceptibility is smaller than that of neutrals and does not deviate by more than one order of magnitude [138] prevents them from having any measurable contribution.

Hence, the dephasing induced by the filament over the whole propagation distance is:

$$\Delta \phi_{XPM}^{probe} = \frac{2\pi}{\lambda_{probe}} \int_{propagation} \Delta n_{XPM}^{probe} dz = \frac{2\pi n_2^{XPM}}{\lambda_{probe}} \int I_{filament} dz$$

(3.6)

The first approximation made here is to neglect group velocity dispersion between the probe and the filament in Argon. Secondly, we neglect the distortions of the spatial and temporal intensity distribution of the filament in considering only the maximal on-axis intensity for the fit. So the resulting inhomogeneities in the birefringence experienced by each spatio-temporal slice of the probe are not taken into account. These two approximations lead to underestimate $n_2^{XPM}$.

A full development of the coupled polarization-dependent equations would be needed to obtain the full expression of the probe dephasing. From the experimental point of view, this corresponds to the pinhole inserted in the beam to select one small, homogeneous region only. Moreover, because of the a priori time dependence of the dephasing, a temporal study of the probe polarization could give a better insight on the temporal dynamics of the filamentation.

Figure 3.6 displays the theoretical on-axis intensity $I_{filament}(z)$ as a function of the propagation distance for several pressure as calculated with the model described in Chapter 1. The induced dephasing between the two polarization axes is then calculated as in Eq.3.6 and shows a linear dependence with the pressure. Indeed, our simulations show that higher pressure results in longer filaments with slightly lower intensity clamping. These effects roughly compensate each other when calculating $\int I_{filament} dz$, so that the dephasing $\frac{2\pi n_2^{XPM}}{\lambda_{probe}} \int I_{filament} dz$ varies like $n_2^{XPM}$, which is proportional to the pressure. The linear tendency is well reproduced by simulations. However, to fit the experimental results, we adopted $n_2^{XPM} = 1.6 10^{-20} cm^2 W^{-1}$, one order of magnitude below expected value of $10^{19} cm^2 W^{-1}$. Further full simulations, including transverse intensity profiles and dispersion effect, are required to provide a quantitative transverse profile of filament-induced birefringence and achieve a better quantitative agreement. As the dephasing depends monotonically on the pressure, any value may be generated by
choosing an adequate Argon pressure. For example, an interpolation of the experimental data presented in Fig. 3.3 suggests that an equivalent $\lambda/4$ plate can be generated for $1.7 \pm 0.1$ bar.

In conclusion, we have demonstrated that laser-generated self-guided filaments can induce substantial birefringence in near-atmospheric pressure gases. An angle of $45^\circ$ between the filament and the probe polarizations allows the realization of Kerr-gates, with an unprecedented switching time ultimately limited by the duration of the filamenting pulse. An optical ultrafast switch could even be initiated remotely by self-guided filaments in the atmosphere [3], even in perturbed conditions [57, 58, 61], opening new perspectives for remote optical ultrafast data transmission and processing, e.g. remote ultrafast optical logical gates.

### 3.2 Interaction between two filaments [139]

In the previous section, the interaction between a weak probe and a filament has been studied. Because the probe is quite weak, it does not induce a change into the propagation of the filament while the filament strongly perturbs the propagation of the probe. In this section, we will study the co-propagation dynamics when the probe is powerful enough to experience filamentation by its own. The interaction which links the two filaments (XPM) is very efficient because the phase matching condition is automatically achieved. Hence the co-propagation of such pulses will lead to strong crossed pulse spatio-temporal deformations. The first part of this section will be dedicated to the description of the experimental setup. In the second part, we will describe the theoretical model which governs the propagation dynamics of such pulses as an extension of the ”single filament” NLSE. Finally, we will analyze both the experimental and theoretical results. In particular, we will see that the two-color filamentation is responsible for a strong enhancement of the spectral broadening, leading to the connection of the two spectra.
Moreover, in appropriate conditions, the two plasma channel can merge, forming a much longer plasma column which could help in the future lightning control experiments. In the temporal domain, we will see that the interaction between the two filaments can provide simultaneously few-cycle pulses at frequencies $\omega$ and $2\omega$. The generation of such pulses could be used in high harmonics experiments to significantly enhance the generation efficiency of attoseconds pulses [140]. The connection of both the two plasma channels and the two spectra led us to name “co-filamentation” the co-propagation of two filaments.

### 3.2.1 Experimental setup

The experimental setup is shown in Fig. 3.7. A Ti:Sa amplifier system (Hydra, Coherent Inc.), delivers 2.5 $mJ$ (stability: 0.5 % rms), 800 $nm$ pulses at a $1 kHz$ repetition rate. The seed pulse is shaped by a Dazzler in order to avoid gain narrowing and consequently, shorten the output pulse down to 30 $fs$. The beam is frequency-doubled to 400 $nm$ using a 0.5 $mm$ thick type I BBO crystal, providing a typical output energy of 150 – 300 $\mu J$. The remaining NIR ($R$, about 1 $mJ$) and the blue ($B$) pulses are separated by a dichroic mirror (DCM). The $R$ polarization is rotated by a zero-order waveplate (WP) and its delay relative to $B$ is adjusted by a delay line, after which the two beams are recombined with another DCM (DCM2). Then, the two collinear beams are focused by a $f = 1 m$ lens in a 2 $m$ long cell filled with 2 bar Argon. We determined the zero-delay between both pulses by optimizing the sum-frequency generation at 266 $nm$ in a 200 $\mu m$ thick BBO crystal. When either of the two pulses propagates into the cell, single filamentation occurs. As expected from the Marburger formula [141] and considering the lens longitudinal chromatic abberation, the $B$ and $R$ filaments start at $z_B = 90 $cm and $z_R = 97 $cm, respectively, $z$ being the propagation distance. The two beams are then collimated at the cell exit by a $f = 1 m$ lens. The core (4 $mm$ width) of the beams are redirected to a spectrometer (Ocean Optics, HR4000, 0.5 $nm$ resolution). Besides, the plasma channels are characterized by side images recorded with a RGB color-frame digital camera (Nikon D80), with a resolution of about 30 $\mu m$. The images were integrated over 3 $s$, corresponding to 3000 laser pulses. We checked that the collected light was completely unpolarized and shows no radial dependence, which excludes both Rayleigh scattering and third-order susceptibility processes, and guarantees that we actually observed plasma fluorescence. To avoid scattered light from either of the $R$ and $B$ filaments, we considered the green layer ($450 – 600$ $nm$) of the RGB pictures, where several fluorescence emission lines of Argon are present, although the main fluorescence signal is in the $650 – 950$ $nm$ range [142].

### 3.2.2 Theory

To get insight into the highly non-linear co-filamentation process beyond the experimentally available data, we extended the numerical model described in Chapter 1 in order to reproduce the actual experimental conditions as closely as possible. In this model, the two pulses are treated separately. This artificial separation is an obvious choice when the two envelope spectra are well separated but can lead to undesirable artefacts, especially when the two spectra overlap [86]. Therefore, we consider two collinearly polarized incident electric fields at
Nonlinear interaction between filaments and ultrashort pulses

Figure 3.7: Experimental setup. **DCM**: Dichroic mirror. **HWP**: Half-waveplate.

\( \lambda_B = 400 \) \( nm \) and \( \lambda_R = 800 \) \( nm \) with cylindrical symmetry around the propagation axis \( z \), written as \( \Re\{\varepsilon_i \exp[i(k_i z - \omega_i t)]\} \), where \( k_i = \frac{2\pi n_i}{\lambda_i} \) and \( \omega_i = \frac{2\pi c}{\lambda_i} \) \((i = B, R)\) are the wave number and the frequency of the carrier waves respectively. The scalar envelopes \( \varepsilon_i(r, t, z) \) are assumed to be slowly varying in time and along \( z \). The only cross-effect between the two beams taken into account is XPM. Therefore, we neglected all phenomena such as \( 2\omega_B \pm \omega_R, 2\omega_R \pm \omega_B \) or higher order phenomena.

The two scalar envelopes evolve according to the coupled propagation equations (1-2) which are the extension of the single pulse model described in Chapter 1:

\[
\partial_z \varepsilon_B = \frac{i}{2k_B} \Delta_{\perp}^{2} \varepsilon_B - i \frac{k_B^2}{2} \partial^2_t \varepsilon_B - i \Delta_{\perp} \partial_t \varepsilon_B + \frac{i k_B}{n_B} \left( n_{2_B} |\varepsilon_B|^2 + n_{cross} |\varepsilon_R|^2 \right) \varepsilon_B \tag{3.7}
\]

\[
\partial_z \varepsilon_R = \frac{i}{2k_R} \Delta_{\perp}^{2} \varepsilon_R - i \frac{k_R^2}{2} \partial^2_t \varepsilon_R + \frac{i k_R}{n_R} \left( n_{2_R} |\varepsilon_R|^2 + n_{cross} |\varepsilon_B|^2 \right) \varepsilon_R \tag{3.8}
\]

where \( t \) refers to the retarded time in the reference frame of the 800 nm pulse: \( t \rightarrow t - \frac{z}{v_{gr}} \).

In comparison with the numerical model presented in Chapter 1, the additional terms on the right-hand side of Eq. 1 account for temporal walkoff due to the group-velocity dispersion of the two envelopes and cross Kerr effects. In (1), \( \Delta_{\perp} = \frac{1}{\nu_B} - \frac{1}{\nu_R} \) is the walkoff constant. All the coefficient values are summarized in Table 3.2.
Nonlinear interaction between filaments and ultrashort pulses

The dynamic of the electric field is coupled with the plasma density $\rho$ because of the multi-photon ionization process. Hence, the plasma density $\rho$ follows the equation:

$$\partial_t \rho = \left( \sum_{l=B,R} \sigma_{K_l}|\varepsilon_l|^{2K_l} \right) \left( 1 - \rho / \rho_{at} \right) + \frac{1}{U} \sum_{l=B,R} \sigma_l \rho |\varepsilon_l|^2 - \alpha \rho^2$$

(3.9)

where $\alpha$ is the recombination time constant.

Initial conditions

We chose an initial plasma density of $10^9 e^{-} \cdot cm^{-3}$ [97]. The input electric field envelopes are modeled in focused geometry by two Gaussian profiles with input power $P_{in}$, as

$$\varepsilon_i(r, t, 0) = \sqrt{\frac{2P_{in}}{\pi \sigma_{r_i}^2}} \cdot \exp \left( -\frac{r^2}{\sigma_{r_i}^2} - \frac{t^2}{\tau_i^2} + i \frac{k_i t^2}{2f_i} \right)$$

(3.10)

$\sigma_{r_i}$ being the intensity quadratic radius, $\tau_i = \Delta t_{FWHM} / (\sqrt{2\ln(2)})$ and $f_i$ the focal length of the $\lambda_i$ pulse respectively. Both the initial delay imposed between the two pulses and the linear chirp of the pulses are set in the frequency space:

$$\varepsilon_B(r, \omega, 0) \rightarrow \varepsilon_B(r, \omega, 0) \cdot \exp(i\omega \Delta t) \cdot \exp(iC_{B\omega}^2 \omega^2)$$

(3.11)

$$\varepsilon_R(r, \omega, 0) \rightarrow \varepsilon_R(r, \omega, 0) \cdot \exp(iC_{R\omega}^2 \omega^2)$$

(3.12)

with $\Delta t$ representing the time delay between the two pulses, $C_B$ ($C_R$) being the linear chirp parameter of the initial $\lambda_B$ ($\lambda_R$) pulse.

<table>
<thead>
<tr>
<th>$k''$ ($fs^2 cm^{-1}$)</th>
<th>$\lambda_B = 400 nm$</th>
<th>$\lambda_R = 800 nm$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$n_2 (m^2 W^{-1})$</td>
<td>$4.9p \times 10^{-23}$ [144]</td>
<td>$3.2p \times 10^{-23}$</td>
</tr>
<tr>
<td>$n_{cross} (m^2 W^{-1})$</td>
<td>$1.p \times 10^{-23}$</td>
<td>$6$</td>
</tr>
<tr>
<td>$K$</td>
<td>$6$</td>
<td>$11$</td>
</tr>
<tr>
<td>$\beta^R (m^{2K-3} W^{1-K})$</td>
<td>$1.95p \times 10^{-88}$</td>
<td>$3.32p \times 10^{-176}$</td>
</tr>
<tr>
<td>$\sigma^R (s^{-1} cm^{2K} W^{-K})$</td>
<td>$2.79p \times 10^{-72}$ [9]</td>
<td>$5.06p \times 10^{-140}$</td>
</tr>
<tr>
<td>$\sigma (1bar) (m^2)$</td>
<td>$2.53 \times 10^{-24}$</td>
<td>$1.01 \times 10^{-23}$</td>
</tr>
<tr>
<td>$\rho_c (m^{-3})$</td>
<td>$6.4 \times 10^{27}$</td>
<td>$1.74 \times 10^{27}$</td>
</tr>
<tr>
<td>$\alpha (m^{-3} s^{-1})$</td>
<td>$7 \times 10^{-13}$ [67]</td>
<td>$1.9 \times 10^{-13} p^{-1}$ [145]</td>
</tr>
<tr>
<td>$\tau (s)$</td>
<td>$1.9 \times 10^{-13} p^{-1}$ [145]</td>
<td>$1 \times 10^{-13} s^{-1}$ [146]</td>
</tr>
</tbody>
</table>

Table 3.2: Physical parameters used in the model (p accounts for the relative gas pressure: $p = \frac{P}{P_{1bar}}$).

The input parameters adopted in this section are summarized in Table 3.3. In particular, the focal length of the plan-concave lens was set slightly longer for the 800 nm (R) pulse than for the 400 nm (B) one to account for longitudinal chromatic aberration.
Nonlinear interaction between filaments and ultrashort pulses

\[ \lambda_B = 400 \text{ nm} \]
\[ \lambda_R = 800 \text{ nm} \]

<table>
<thead>
<tr>
<th></th>
<th>(\lambda_B = 400 \text{ nm})</th>
<th>(\lambda_R = 800 \text{ nm})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Energy (mJ)</td>
<td>0.2</td>
<td>1</td>
</tr>
<tr>
<td>(\Delta t_{FWHM}) (fs)</td>
<td>30</td>
<td>30</td>
</tr>
<tr>
<td>(\sigma_r) (mm)</td>
<td>6</td>
<td>6</td>
</tr>
<tr>
<td>Chirp (fs²)</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>(f) (m)</td>
<td>1</td>
<td>1.04</td>
</tr>
<tr>
<td>Pressure (bar)</td>
<td>1</td>
<td></td>
</tr>
</tbody>
</table>

Table 3.3: Input parameters used as initial condition of the simulations presented in this section.

3.2.3 Results and discussion

Spatio-temporal dynamics of co-filamentation

To precisely identify the contribution of co-filamentation, we compared the results of numerical simulations of both the independent filamentation of 400 nm and 800 nm with the case where the two pulses co-propagate, with the same initial conditions. Fig. 3.8 displays the effect of co-filamentation on both the quadratic radius and the intensity in the center of the beams, as a function of the propagation distance. While \(\lambda_B\) is little affected by the co-filamentation, \(\lambda_R\) is refocused twice by cross-Kerr focusing. In this process, the photon bath [58, 146] of \(\lambda_R\) significantly contributes, through a \(\chi^{(3)}\) process, to the refocusing of the co-propagating \(\lambda_B\) pulse. In contrast to \(\lambda_B\), \(\lambda_R\) is moderately affected by the co-filamentation. This is due to the fact that the \(\lambda_B\) transverse dimension is larger than the \(\lambda_R\) one almost all along the propagation (typically 100 \(\mu m\) vs 25 \(\mu m\)), so that the nonlinear cross-effects averaged over the whole \(\lambda_R\) cross-section remain small. The most significant cross-effect experienced by the \(\lambda_R\) pulse is a defocusing induced by the plasma generated by \(\lambda_B\) (see Fig.3.9). The numerical simulations also show that the clamped intensity of both pulses is almost unaffected by co-filamentation and amounts to \(1 \cdot 10^{13} \text{ W.cm}^{-2}\) for \(\lambda_B\) and \(6 \cdot 10^{13} \text{ W.cm}^{-2}\) for \(\lambda_R\) over a few centimeters and that the optical fluence reaches comparable value of about \(1 \text{ J.cm}^{-2}\) for both pulses.

Figure 3.10 (resp. Fig. 3.11) represent \(\lambda_B\) (resp. \(\lambda_R\)) intensity as a function of time and radial distance, for several propagation distances. In both figures, the first column corresponds to the propagation dynamics of one-color filamentation, while the second one shows the result of the co-filamentation process. The initial delay between the two pulses is set at 0 fs (i.e. the two pulses perfectly overlap at \(z = 1 \text{ m}\) in the linear regime).

Because the two filaments are not superimposed longitudinally (\(\lambda_B\) being slightly before \(\lambda_R\)), the \(\lambda_B\) filament generation is helped with the quite intense \(\lambda_R\) energy bath. Hence Kerr lens is more intense. Conversely, \(\lambda_R\) being transversally large compared to the \(\lambda_B\) filament (typically 25 \(\mu m\) (resp. 100 \(\mu m\)) for \(\lambda_B\) (resp. \(\lambda_R\))), the nonlinear phase shift experienced by the total red pulse remains negligible and the \(\lambda_R\) global spectrum is not significantly broadened. However, when the \(\lambda_B\) intensity reaches the multiphotons ionization threshold, the NIR pulse core is both absorbed and diffracted due to multiphoton absorption and ionization respectively (see Fig.3.9(b)). The cross-Kerr lens generated by the blue filament focuses \(\lambda_R\) earlier. When the \(\lambda_R\) filamentation occurs, the spiky blue pulse experiences more nonlinear phase shift due to the
Figure 3.8: (a,c) Quadratic radii of both pulses (blue: 400 nm filament. Red: 800 nm filament) as a function of propagation distance when (a) the two pulses co-propagate and (c) propagating separately. During the filamentation process, B (resp. R) radius is about 25 µm (resp. 75 µm). When the two pulses co-propagate, a second focusing cycle is observed at z = 110 cm. (b,d) Intensity of both pulses centers. (Blue: B intensity. Red: R intensity) when (b) the two pulses co-propagate and (d) propagating separately. During the filamentation process, B (resp. R) is clamped at about 90 TW · cm⁻² (resp. 50 TW · cm⁻²). B intensity increases as R experiences filamentation due to the cross-Kerr focusing.
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Figure 3.9: a) 400 nm and b) 800 nm fluence as a function of the propagation distance. The solid lines depict the quadratic diameter of the pulse fluence during the propagation.

Combined effects of XPM and multiphoton ionization blue shift, which leads to a broadening enhancement (Fig. 3.12). The shape of this broadening depends on the delay between the two pulses, which determines which of the spikes of the B pulse experiences the R-induced phase shift.

Ultra-broadband continuum generation: spectral connection between co-propagating filaments

When the beams propagate separately, the spectral broadening is only due to SPM and, to a lesser extend, to plasma-induced nonlinear phase shift. In the case of co-filamentation, addi-
Figure 3.10: One or two color filamentation dynamic for the 400 nm pulse. Left: 400 nm intensity for several distances when propagating alone. Right: 400 nm intensity for several distances when the NIR filament copropagates.

Tional XPM-induced nonlinear phase shift increases the spectral width of the continuum. The blue curve in Fig. 3.13(b) depicts the blue filament theoretical spectrum when it propagates alone in the cell. When the red filament copropagates with the blue one, a fine delay tuning allows to control the shape of the blue spectrum output.

Figure 3.13(a) shows experimental spectra around 400 nm as a function of the delay between the pulses. The numerical results agree quite well with the experimental ones, although the latter are not as broad as the former. This discrepancy is due to the fact that the experimental data integrate the spectrum over a 4 mm wide region, while the numerical results consider the spectrum in the filament center, where the broadening is strongest. The presence of a pre-pulse in the experiment [32] and an unperfect transverse intensity profile may also contribute to this
Figur3.11: One or two color filamentation dynamic for the NIR pulse. Left: 800 nm intensity for several distances when propagating alone. Right: 800 nm intensity for several distances when the blue filament copropagates.
Figure 3.12: Pulses spectra as a function of the distance when they propagate independently (a,b) and in the co-filamentation regime (c,d). The co-propagation of the two pulses induces a strong spectral broadening due to XPM.

filaments propagating alone. The spectrum does not only exhibit a broader continuum at both the R and B wavelengths, but it also bridges those two continua into a single supercontinuum spanning over two octaves. Figure 3.14 also displays the electric field (b) and the associated intensity (c) if one assumes a perfect re-compression of the whole spectrum, yielding a pulse as short as 1.4 fs FWHM.

Towards the simultaneous generation of few-cycle pulses [147]

In the previous section, we saw that the interaction between two filaments leads to an enhancement of the spectral broadening. However, because of the group velocity dispersion, the pulses will not be a priori Fourier transform limited and consequently, a temporal compression is needed to obtain ultrashort laser pulses.

In this section, we will see that the interaction of the two filaments can simultaneously generate two blue and red few-cycle pulses without any compression.

Increasing interest has recently been devoted to the generation of single attosecond pulse generation because they allow to probe phenomena such as coherent molecular dynamics or Rydberg orbital motion with attosecond time resolution [29, 148, 149]. The generation of at-
to second pulses require few- or even single-cycle driving pulses, raising, in turn a strong attention to few-cycle pulses (FCP) generation [34–36, 150].

Filamentation provides an efficient way to produce such FCP [31, 151, 152]. So far, two main techniques based on filamentation have been proposed in order to generate FCP .

The first approach consists in stopping the filamentation by a gas pressure gradient, before pulse splitting occurs. But this approach requires a complex gas cell design and a careful control of the longitudinal location of filamentation [39]. In the second approach, filaments generate a broad spectrum by self-phase modulation (SPM). This continuum is then temporally compressed by chirped mirrors or a pulse shaper [31]. The complexity of this approach stems from the recompression stage, which requires both careful design and alignment. However, a critical point is also the spectral width of the continuum. The use of a dual-color pulse can improve this bandwidth. Recently, numerical simulations suggested that a weak seed pulse at frequency $\omega_s$ co-propagating with a filaments at $\omega_0$, can generate an ultra-broad continuum at $\omega_{4WM} = 2\omega_0 - \omega_s$ through four-wave mixing (4WM) and produce a FCP even without any post-compression stage [153]. The induced light bullet can then propagate over about 0.25 m, much longer than expected by considering the group-velocity dispersion.

Besides enhancing the spectral broadening, the use of a two-color pair of ultrashort pulses at frequencies $\omega$ and $2\omega$ breaks the temporal symmetry of the electric field. Therefore, the emission of attosecond pulses in the cutoff photon energy region takes place every full cycle instead of every half-cycle, so that the duration of the driving pulse can be doubled and extended to the multi-cycle regime, instead of being confined to single-cycle pulses [140,148,154–157].

Hence, the simultaneous generation of a two-color pair of ultrabroadband pulses at both $\omega$ and $2\omega$ would simultaneously help both stages of the attosecond pulse generation: the generation of the FCP and the subsequent production of single attosecond pulses in the higher harmonics. In this section, we numerically show that the co-filamentation of two laser pulses

**Figure 3.13:** 400 nm pulse spectrum as function of 400 nm- 800 nm relative delay. The time delay is positive when the NIR pulse R is launched after the blue B. a) shows the experimental results when b) the theoretical ones.
Figure 3.14: a) Spectral broadening enhancement (experimental) due to the co-propagation of two filaments. Electric field (a) and related intensity (c) associated with the supercontinuum depicted in a) if assuming a perfect recompression.
at respectively $\lambda_R = 800 \, nm$ and $\lambda_B = 400 \, nm$ in Argon indeed generates a pair of intense, sub-6-cycle pulses in a two-color field, meeting the requirements for attosecond pulse generation. Moreover, contrary to existing experimental schemes, this two-color pair of pulses is obtained without any temporal post-compression stage nor complex gas cell design, leading the way to a practical experimental implementation.

The co-propagation of $B$ and $R$ filaments does not fully prevent pulse splitting (Fig. 3.15). However, its effect can be drastically counteracted resulting in both $R$ and $B$ ultrashort pulses (Fig. 3.16) if the initial time-delay is correctly adjusted ($\Delta t = 0$). As shown in Fig. 3.16, the co-filamentation strongly influences the temporal shape of $B$. The independent propagation of $B$ gives rise to multiple pulse splitting, resulting in a series of sharp spikes spanning over 100 $fs$, which would fully prevent single attosecond pulse generation. In contrast, cross-Kerr focusing, as well as defocusing and absorption of each pulse by the plasma generated by the other one, partially blocks pulse splitting, generating a single 6.5 $fs$ long pulse (4.9 cycles) with no quadratic phase dependency. In the temporal region of high intensity, the phase exhibits a linear temporal dependency which indicates a central frequency shift of $B$ (the negative slope indicates a central frequency blue shifting as can be noticed in Fig. 3.12). Besides this slight blue-shift, the spectrum of the continuum is widely broadened as compared with the case of the $B$ filament propagating alone (Fig. 3.12). Similar to the case of the spatial domain, co-filamentation affects $R$ marginally, although it typically reduces the pedestal intensity by 2 and increase the contrast ratio between the fluence of the almost-FCP ($15 \, fs$, i.e. 5.6 cycles) and its pedestal by 2.5.

The space-time plots of Fig. 3.15 give more insight into the co-filamentation spatiotemporal dynamics. The drastic reduction of pulse splitting as well as the cross-Kerr focusing appear clearly. After $B$ has experienced multiple pulse splitting, the $R$-induced plasma diffracts and absorbs all the $B$ spikes, which temporally occur after $R$). Simultaneously, the (cross) Kerr effect focuses both the $B$ and $R$ energy reservoirs located around the filaments. Moreover, the
Figure 3.16: Temporal intensity distribution (solid line) and temporal phase (dashed line) of 400 nm and 800 nm pulses after co-filamentation (a,b) and after independent filamentation (c,d) at \( z = 1.5 \) m, i.e. 50 cm after the (co)filamentation process.
effect of co-filamentation on \( R \) can be noticed in Fig. 3.15(b) at the distance \( z = 94 \, cm \), where the plasma generated by \( B \) defocuses the trailing edge of \( R \). Without any post-compression, \( R \) is composed of only 5.6 cycles (15 \( fs \), while \( B \) contains about 4.9 cycles (6.5 \( fs \)) at \( z = 150 \, cm \) with a contrast 3.5, 50 \( cm \) after the filamentation process. At this distance, \( R \) (resp. \( B \)) intensity is as high as 1 \( TW \cdot cm^{-2} \) (resp. 0.3 \( TW \cdot cm^{-2} \)) corresponding to a bullet energy of 160 \( \mu J \) (resp. 13 \( \mu J \)) integrated over 2 \( mm \) and considering only the main temporal pulse. Moreover, the extremely short FWHM time duration of the two light bullets remains stable over more than 20 \( cm \) as expected for temporal soliton-like structures. It may be noted that the above results have been recorded in the center of the beam, although none of the two pulses is transversely inhomogeneous. However, real experiments compensate the inhomogeneities and select the shortest and spectrally broadest regions in the center of the beam using a pinhole. Therefore, we expect that our calculations are indeed representative of future experiments. These experiments could greatly benefit of the self-aligned character of the proposed technique, as well as the fact that it requires no post-compression. Such advantages, together with its reasonable energy conversion efficiency, make our approach experimentally very attractive as compared with techniques providing a higher yield, but at the cost of more elaborate setups such as hollow waveguides and chirp mirror recompression [158].

The final FWHM duration of \( B \) is highly sensitive to the initial delay between the two pulses. Figure 3.17(a) displays the FWHM temporal duration of the \( B \) pulse at \( z = 150 \, cm \) after the co-filamentation process. A small detuning of the delay leads to dramatic increase of FWHM duration and to a decrease of the contrast between the main peak and the bath (Fig.3.17(c)). For adequate delays, the interaction with the \( R \) pulse leads to the inhibition of the pulse splitting which occurs during the filamentation of the \( B \) pulse. On the other hand, when the delay is detuned, the pulse splitting is enhanced which results in a longer pulse composed of several sharp peaks. The \( R \) duration is less sensitive to the delay tuning. A change of about 20 \( \% \) is expected concerning the FWHM pulse duration of \( R \) when tuning the delay (Fig. 3.17(b)). However the contrast between the intensity of the \( R \) pulse and its pedestal can be increase up to a factor 3 when adjusting the initial delay (Fig. 3.17 (d)).

As a conclusion, we have numerically demonstrated a compression-free method to simultaneously generate two sub-6-cycle pulses with sub-Joule energy. The co-filamentation of two pulses, and the associated cross-Kerr focusing, drastically reduces the pulse splitting, resulting in almost few-cycle pulses at both \( \omega \) and \( 2\omega \). Such doublet is of particular interest as a pair of driving pulse for the generation of attosecond pulses. The pretty high, almost not record-breaking energy conversion efficiency of this method, combined with the fact that it is self-aligned and self-recompressed, makes it attractive for practical experiments.

**Generation of long connected plasma channels**

In this section, we focus our attention on the medium ionization induced by co-filamentation.

In the case of “classical” filamentation (involving only one pulse), when the laser intensity exceeds a few \( 10^{12} \, W.cm^{-2} \), Argon ionizes. The resulting plasma stabilizes the filaments in a dynamic balance with the Kerr effect. Therefore, the length of the plasma channel is representative of that of the filaments themselves [159]. Moreover, the plasma decreases the local medium resistivity, which is of high importance to control high-voltage discharges [56] or light-
Nonlinear interaction between filaments and ultrashort pulses

Figure 3.17: Blue (resp. red) pulse FWHM temporal duration (a) (resp. (b)) and the associated contrast ratio between the main peak and the bath (c,d) at $z = 150$ cm after the co-filamentation process. Negative (resp. positive) delay corresponds to the situation where B s launched before (resp. after) R. A delay 0 means that the two pulses arrive simultaneously at the focus point in the linear regime.

Long, highly conductive plasma “wires” are therefore highly desirable in order to increase the triggering and guiding efficiency of the filaments. Figure 3.18(a) depicts the experimentally measured plasma density as a function of the propagation distance, for several delays between the R and B filaments. As long as the pulses do not temporally overlap, the resulting plasma density is the sum of the contributions of the two pulses propagating individually, with two plasma channels of 2.5 cm length, separated by a 2 cm long non-ionized (therefore, isolating) region. When the pulses overlap, the two plasma column do not only connect, but the gap between them is filled by a continuous plasma channel. In other words, co-filamentation bridges the individual plasma channels into a 9 cm long one, corresponding to a 3.6-fold increase of the length as compared with the individual filaments. It is indeed remarkable that this bridging is obtained by the addition of the B pulse, which accounts for only 13 % of the overall energy involved in the experiment. The same lever effect is observed in the numerical simulations (Fig. 3.18(b)). It results in a decrease of the filament resistivity between $z_1 = 0.9$ m and $z_2 = 1.05$ m, which is proportional to $\int_{z_1}^{z_2} 1/\rho(z) dz$. The co-filamentation reduces this resistivity by a factor 1.7, as compared to the independent propagation of the individual pulses, i.e. when they do not temporally overlap. Such situation contrasts with the concatenation of two plasma channels generated by two collinear 800 nm pulses of similar 4 mJ pulse energy [160]. In the latter case, adjacent filaments are connected, but no non-ionized gap can be bridged between them. The doubling of the plasma channel length appears in the latter case as a straightforward effect of doubling the launched energy, rather than of a non-linear interaction between two co-filamenting pulses.

Partial temporal overlap between the two pulses results in intermediate results. The addition of R behaves as a supplementary energy reservoir [161] and yields an additional cross-Kerr focusing which shifts the onset of the B filaments toward the laser source. Reciprocally, B contributes to focusing R, but its plasma also defocuses and absorbs R. The outcome of these opposite effects depends on the sign of the delay between the pulses. If R is launched after
**Figure 3.18:** a) Experimental plasma channel length for several distances. Blue: plasma channel generated only by $B$. Red: plasma channel generated only by $R$. Green: plasma channel generated by the two pulses. For well adjusted delay, the two plasma channel are concatenated and form alonger plasma channel without any non-ionized region. Blue: $B$ induced plasma channel. Red: $R$ induced plasma channel. The other plots depicts the plasma density when $B$ and $R$ co-propagates for several input delays. b) Theoretical plasma channel.

$B$, the plasma left behind by $B$ tends to defocus the $R$ pulse, so that the $R$ filament starts later and has a lower plasma density. On the opposite, if $R$ is launched before $B$, the cross-Kerr focusing dominates. The resulting plasma channel is more intense and shifts upstream. Therefore, the plasma channel length, density and location can be controlled by adjusting the time delay between the two pulses.

### 3.3 Conclusion

In this Chapter, we have discussed the interaction between a filament and another electric field. An intense electric field can stretch the spherical electron cloud of atoms into an ellipsoid elongated along the electric field polarization direction, resulting in an induced birefringence. Laser-generated self-guided filaments can induce substantial birefringence in near-atmospheric pressure gases. An angle of $45^\circ$ between the filament and the probe polarizations allows the realization of Kerr gates, with an unprecedented switching time ultimately limited by the duration of the filamenting pulse. An optical ultrafast switch could even be initiated remotely.
by selfguided filaments in the atmosphere, even in perturbed conditions, opening new perspectives for remote optical ultrafast data transmission and processing, e.g. remote ultrafast optical logical gates.

The interaction between two filaments also results in a strong mutual influence, a process that we named co-filamentation. The unique interaction between these two self-channeling structure is very efficient because the phase matching condition is automatically fulfilled. It results in the merging of both the plasma channels and the spectra continuum. Moreover, in appropriate conditions (delay, chirp, energy), the propagation dynamics can lead to the simultaneous generation of few-cycle pulses without any post-compression stage. Such doublet is of particular interest as a pair of driving pulse for the generation of attosecond pulses. The pretty high, almost not record-breaking energy conversion efficiency of this method, combined with the fact that it is self-aligned and self-recompressed, makes it attractive for practical experiments.
Chapter 4

Alisé facility: towards a multi-TW white-light LIDAR [162]

In the previous Chapters, the nonlinear propagation of GW laser pulses has been studied. Hence the situations described so far relate to pulses having an incident power around $P_{cr}$, the threshold power for critical Kerr self-focusing. Ten years ago, the emergence of ultrashort lasers with peak powers surpassing by several orders of magnitude the critical power ($P \gg P_{cr}$) had raised a lot of interrogations about the scalability for TW lasers, especially because the numerical simulation of such kind of laser propagation needs massive parallel computing [89] only accessible to a few research groups. However, during this last ten years, a lot of theoretical and experimental results have, to a great extent, answered to the main questions which have been raised. In particular, it has been shown that increasing the peak power does not lead to a catastrophic collapse of the pulse nor to a more energetic filament. On the contrary, the pulse breaks up into several filaments of equal energy: this regime is called multiple filamentation regime [7]. It mainly occurs because of two main physical mechanisms. The first mechanism was proposed by Bespalov and Talanov [163]. Multiple filamentation occurs because of small random perturbations of the medium and/or in the input beam profile. These perturbations grow around the sharp gradients of the beam profile according to modulational instabilities and lead to the formation of filaments. A few years ago, an alternative deterministic theory was proposed by Fibich and Ilan [75]. The breakup into several filaments has been attributed to vectorial effects which break the axial symmetry while inducing a preferred direction in the transverse plane (the direction of input beam polarization). This symmetry breakup then lead to the multiple filamentation regime.

In this Chapter, we investigate the scalability of this propagation further these energies and powers by launching multi-Joule, multi-Terawatt laser pulses delivered in the atmosphere by the Alise beamline of the CEA-CESTA. Propagation of pulses at this extreme energy level has not been investigated so far neither experimentally nor theoretically. As it happened 10 years ago for TW pulses, nobody is able to predict what will happen at this energy level during the propagation and the understanding of the propagation dynamics is highly challenging.


4.1 The Alisé facility

Alisé is a high power Nd:glass laser chain providing an energy up to 200 $J$ at $\lambda_0 = 1053 \text{ nm}$, with a FWHM spectral width of 3 $nm$. The beam is mainly used for studies on laser physics and for ultra-high intensity interaction with solids, gases, and plasmas both within the CEA and through collaborations with the international scientific community. The laser pulses are generated, amplified and shaped before being directed to the laser hall. The facility is composed of three different laser sources depending on the requirements of the experimenters: The ”typical LMJ” (Laser MégaJoule) source, the CADDO source (Cavity Dumping Dove Oscillator) and the stretched beam for chirped pulse amplification (CPA). In the CPA configuration, which we used in our experiments, the Alisé laser amplification chain makes it possible to raise the beam energy from a few mJ to over 200 $J$ with an output beam of 90 $mm$ in diameter. The architecture of the Alisé laser amplification chain includes: three 50 mm diameter rod amplifiers, two 94 $mm$ diameter disk amplifiers, one 150 $mm$ diameter disk amplifier, and five spatial filters enabling filtering of the beam and adapting its diameter for the various amplifiers (Fig. 4.1). The firing frequency is 1 shot per hour. For laser beam diameters of 30 to 37 $mm$ and energies of 3 to 5 $J$, firing frequency is increased to 1 shot every 15 minutes. The minimal laser output pulse duration is about 500 $fs$, limited by the spectral gain narrowing of the phosphate Nd:glass.

4.2 Experimental setup

We investigated the vertical propagation in air of pulses from the Alisé laser facility. Alisé was used in its chirped pulse amplification (CPA) configuration. The pulse energy was limited to 26 $J$ energy by the damage threshold of the atmospheric pressure compressor. During the experiment, the pulse duration was varied from 550 $fs$ to 65 $ps$ FWHM. The corresponding peak power amounts to up to 32 $TW$ peak power, corresponding to about 5,000 times the critical power for self-focusing ($P_{cr} = 6 \text{ GW}$ at 1053 nm, considering $n_2 \simeq 3 - 4 \times 10^{-19} \text{ cm}^2/\text{W}$ in the air [164]). The beam was emitted vertically into the atmosphere, either collimated (natural divergence of 10 $\mu rad$), or focused through lenses ($f = 16 \text{ m}$ or $f = 300 \text{ m}$) installed at the exit of the grating compressor. Laser diagnostics included a beam profile analyzer, as well as a single-shot autocorrelator and a streak camera, which yielded the duration of the shorter and longer pulses respectively. A 10 $mJ$, 2 $ns$ frequency-doubled Nd:YAG laser, collinear with the Alisé beam, was used as a reference for a quantitative estimation of the conversion efficiency into the white-light continuum (Fig. 4.2). The backscattered white light signal was detected by a slightly off-axis (35 cm) Lidar system consisting of a 20 cm telescope equipped with detectors sensitive in three spectral ranges in the visible and near-infrared (Fig. 4.3). Further spectral selectivity was achieved by using bandpass filters. Simultaneously, the beam was imaged from the side, from an off-axis distance of 5 to 30 $m$, by a color-frame, digital CCD camera (Nikon D70) providing a 1.5$^\circ$ field of view, and a resolution of $5 \times 10^{-4}$ degrees. Alternatively, the beam profile was recorded on photosensitive paper after 11 $m$ propagation.
Figure 4.1: a) Schematic of the Alise facility. 4 amplification stages are used to obtain 30 J per pulse. The pulse is then temporally re-compressed in a separate room. The final maximal specifications of Alise are 200 J, 500 fs, 32 TW. The laser is then launched into the sky. A driving Nd:YAG is used to align the Alise laser. b) Picture of the amplification room (after CEA documents).
**Figure 4.2:** Beam of the 10 mJ, 2 ns 532 nm driving Nd:YAG used for the detection alignment and LIDAR intensity calibration.

**Figure 4.3:** Experimental setup for Lidar measurement.
4.3 Results and discussion

4.3.1 Observation of multiple filamentation in multi-TW laser beams

Once pulses are launched in the air, Kerr effect is initiated in the beam. For the ultra high powers at play in our experiment \((P \gg 100 \, P_{cr})\), multi-filamentation occurs through modulational instabilities which break up the beam into periodic cells over very short propagation lengths \(z_{fil} \sim 2 \frac{P_{cr}}{\Delta n I_0} \approx 1 - 3 \, m\) for an incident intensity \(I_0 \approx 4 - 6 \times 10^{11} \, W/cm^2\). Hot spots can be observed on the beam profile recorded on photosensitive paper after \(11 \, m\) propagation (Fig. 4.4), which correspond to the onset of mature individual filaments (Fig. 4.4(d)). These high intensity region are linked by lower intensity bridges, which appear as straight lines in Fig. 4.4(d), and generate a typical honeycomb beam structure \([85, 165, 166]\). The high intensity of the hot spots is confirmed by their ability to locally ablate the paper surface within a single shot. Calibrated ablation tests showed that the intensity within the filaments is about one order higher than the clamped intensity of \(5 \times 10^{13} \, W/cm^2\) observed in sub-Joule beams at 800 nm \([47]\). This finding suggests that even at multi-Joule energies, the filamentation process is governed by the local dynamic balance between Kerr effect and plasma defocusing within the beam profile. Hundreds of filaments are observed within the beam profile unless the beam is focused too strongly (Fig. 4.4(c)). We counted typically one filament for each \(3.5 \text{ to } 7.5 \, P_{cr}\), very close to the ratio observed at lower power with the Teramobile \((5P_{cr}/\text{filament})\) \([58]\), although the latter experiments were performed at a wavelength of 800 nm instead of 1.06 \(\mu\)m. The occurrence of self-guided filaments shows that, as is the case for lower pulse energy, Kerr lens self-focusing does not cause the beam to collapse, but instead promotes a local dynamic balance with defocusing induced by the electron plasma generated at the non-linear focus of each self-focusing cell. This allows the beam to propagate collimated at long
distances in spite of strong self-focusing. Filaments generated in the beam emit the white-light continuum forward as a "white-light laser" [167], which was clearly visible by naked eye, propagating to high altitudes in the zenith direction as a collimated beam, comparable with similar observations at lower power with the Teramobile.

To efficiently exploit the possibilities opened by multi-Joule pulses in the atmosphere, one critically needs to control their propagation. For this reason, we investigated the effect of the pulse duration on the filament location (onset, and filamentation length). This was performed by adjusting the laser chirp [7]. The effect of chirp on filamentation clearly appears when comparing images of the beam in the visible spectral region (Fig. 4.5). From the images, we

**Figure 4.5:** Chirp dependence of the filament onset. (a) Side image of the beam. Signal corresponding to larger initial chirps, (lower peak powers), rise more slowly but over longer distances, showing that the white-light is still generated at higher altitudes, although filamentation is weaker in this case. (b) Observation geometry.
evaluated the intensity profile of white-light as a function of altitude. The reference YAG laser, which propagates linearly, was used as a reference for intensity normalization. This procedure allowed us to get rid of the atmospheric variability and of slight day-to-day differences in the detection alignment. In this analysis, an increasing normalized signal is the signature for white-light generation, and therefore for filamentation. We observe white-light already at the bottom of the image (19 m) for all chirps, although larger chirps (i.e. lower peak powers) yield less efficient white-light conversion per unit length. The shortest pulses (570 fs, i.e. 32 TW) yield shorter filamentation (filament end at 100 m) while 2.1 ps pulses (9 TW) push the filamentation end 350 m away from the laser source (Fig. 4.6). Such a filament length is remarkable: Filaments observed at lower energy levels, although they are currently restricted to some tens of meters in length may be generated at km-range altitudes. The observed altitude dependence on the pulse duration of the laser beam shows that filamentation of ultra-high power, multi-joule laser pulses can be controlled remotely by changing the laser parameters as is currently performed on smaller laser classes.

4.3.2 LIDAR observation for multi-filamentation characterization

We used a LIDAR configuration to analyze the laser source from the signal received by our detection telescope. This "inverse problem" method allows to study the propagation of the Alisé beam through the atmosphere over its whole propagation length at once. However, the highly
**Figure 4.7:** Single-shot white-light Lidar signal in the $300 - 470 \text{ nm}$ spectral region, displaying signal up to $20 \text{ km}$ in spite of a cloud layer (cirrus) around $10 \text{ km}$. 

![Graph showing Lidar signal with distance and wavelength features](image-url)
The nonlinear nature of the propagation of such laser and the variability of the atmosphere composition between two consecutive shots (1 shot per hour) make the LIDAR signal interpretation very challenging. This complexity leads us to make a strong approximation by interpreting the LIDAR signal in terms of equivalent linear LIDAR signal, i.e. by considering the LIDAR signal as those of a linear laser giving the same LIDAR signal.

The laser supercontinuum was observed over the whole visible spectrum, from 300 nm to 850 nm, showing it is extremely broad. Figure 4.7 displays the white-light Lidar signal detected in the 300–475 nm spectral region, as a function of altitude. Although the acquisition is single shot, the signal can be clearly observed up to 20 km, in the stratosphere. This is all the more remarkable that the considered spectral region of the visible is more than 600 nm away from the fundamental laser wavelength. This spectacular result constitutes the most powerful Lidar signal acquired so far, and definitely assesses the capability of 30 TW pulses to propagate over kilometer-range distances without diverging.

One of the goals of our experiment was to determine a quantitative energy spectrum of Alisé after the propagation in three main spectral regions: [400, 500, 650 nm]. In that purpose, we acquired before each shot a LIDAR signal of the pilot Nd:YAG in order to be able to precisely determine the backscattering coefficient and the receiver telescope parameters such as the inclination angle, and the distance from the laser. Figure 4.8 displays the fit provided by a Levenberg-Marquardt method [168] of an experimental LIDAR signal with our numerical model (see Annex 1), giving the closest (in the sense of Nonlinear Least-Squares [169]) LIDAR signal shape and laser energy. The fit consequently confirms the distance between the telescope and the laser \((d_0 = 35 \text{ cm})\) and the angle between the laser propagation direction and the optical axis of the telescope \((\delta = 3.5 \text{ mrad})\). The fit also allows, for each shot, to quantitatively calibrate the backscattering coefficient \(\beta\). In particular, according to Eq. 17 (Annex 1), one have to know the aerosol size distribution to retrieve the backscattering coefficient \(\beta\). We use a typical Deirmenjian "Haze M" distribution \([175]\) \(n(r) = a r^\alpha e^{-b r^\gamma}\) representing coastal conditions (the CEA laboratory is located only about 20 km away of the Atlantic ocean). Moreover, we assume that this distribution is independent from altitude within the considered range \([0 – 1000 \text{ m}]\).

The driving Nd:YAG allows to calibrate the total density of aerosols \(N = \frac{2 \pi}{\lambda} \frac{a^{\alpha+1}}{\Gamma(\alpha+1)}\) present in the atmosphere. Consequently, with an aerosol density \(N = 350 \text{ cm}^{-3}\) retrieved from the LIDAR signal of the calibration laser (for the results described in this Chapter), this distribution reads:

\[
n(r) = 1.9 \times 10^5 r e^{-8.94 r^{0.5}}
\]

where \(n\) is in \(\text{cm}^{-3} \mu m^{-1}\) and \(r\) in \(\mu m\) (see Fig. 4.9).

We are then able to calculate \(\beta\) for any wavelength \(\lambda\). Figure 4.10 displays the backscattering coefficient \(\beta\) as a function of the incident wavelength.

These parameters were then inserted into our model to fit the Alisé signals in order to give an estimation of the equivalent energy at each spectral region of the continuum. Figure 4.11 displays typical LIDAR signals at 500 nm for two different initial pulse durations (5 ps and 15 ps) and their respective fits.

As it is depicted in Fig. 4.12, inverting the LIDAR equation allows to retrieve the equivalent energy in the corresponding spectral region of the continuum as a function of the altitude.
Figure 4.8: Numerical fit of the driving Nd:YAG LIDAR signal.

Figure 4.9: "Haze M" size distribution function used in the integration of the Mie function.
Figure 4.10: Backscattering coefficient $\beta$ as a function of the incident wavelength.

Figure 4.11: Example of numerical fits of Alisé LIDAR signals at 500 nm for two different chirps.
Figure 4.12: Energy of the white light as a function of the distance for several initial pulse durations.
Figure 4.13 displays the energy averaged over 500 m in three spectral regions as a function of the initial pulse duration. The analysis shows that the chirp almost does not affect the ratios between different wavelengths, i.e. the shape of the spectrum of the continuum generated in the filaments. Moreover, signal detected at a given wavelength (hence, the generation efficiency of the white-light at this wavelength, at the fixed incident pulse energy of 15 J) is inversely proportional to the pulse duration. We estimate the absolute value of the signal at 650 nm for the shorter pulses (520 fs) to be 2.2 mJ, corresponding to a conversion efficiency of $\sim 1.5 \times 10^{-5} \text{nm}^{-1}$ for 31 TW laser pulses. Considering the typical width of the continuum and its expected exponential decay away from the fundamental wavelength [24, 167], this figure leads us to estimate that the overall conversion efficiency from the fundamental into the continuum amounts to a few percents. This conversion efficiency is smaller than may have been expected from extrapolations based on previous measurements at 800 nm at lower power and energy [167]. Although longer fundamental wavelengths could have been expected to yield a more efficient broadening [30, 170], this observation can be understood when considering that the intensity within the filaments is clamped and that their number, which is proportional to the power, does not influence much the shape of the white-light spectrum [171]. Spectral broadening is thus governed by the time gradients of the intensity through nonlinear phase variations. As detailed in Chapter 1, these gradients are sharper for shorter pulses and therefore yield a more efficient broadening [172]. Hence, further optimization of the white-light generation would require one to increase the peak power by shortening the laser pulses, even at the cost of a reduced pulse energy.
4.4 Conclusion

As a conclusion, we have shown that ultra-intense laser beams up to 30 TW, 20 J do not collapse when propagating in the atmosphere, but instead generate multiple (up to 400) filaments through processes remarkably similar to those observed for sub-Joule pulses, excepted for a higher intensity within them. Filamentation can be controlled by varying the duration and energy of the pulses. Although the generation efficiency of the associated white-light supercontinuum, which is inversely proportional to the pulse duration, is restricted to a few percent, the white-light propagates up to the stratosphere, i.e. beyond 20 km, constituting the highest power “white-light laser” to date. This intense white-light source may be further enhanced by using shorter pulses. These results are encouraging for the use of multi-joule, ultrashort laser pulses in both future nonlinear white-light Lidars or applications requiring the remote delivery of high intensities, such as lightning control.
Chapter 5

Conclusion and Outlook

In this Thesis, the filamentation process has been studied in gas both numerically and experimentally. First, we have derived, from the Maxwell’s equations, the Nonlinear Schrödinger equation (NLSE) which drives the propagation dynamics of high power ultrafast laser pulses in the special case of paraxial and slowly varying envelope equation. It allowed the implementation of a numerical model resolving the NLSE in cylindrical symmetry, providing a better understanding of the nonlinear propagation dynamics. In particular, we have identified the global dependence of the output pulse characteristics (spectral width, intensity, length and density of the plasma channel) on the initial conditions (energy, duration, chirp, focusing, and gas pressure).

In Chapter 2, we have investigated the statistical properties of ultrashort pulses propagating in chaotic media. First, we have demonstrated that during the linear propagation through a turbulent medium (such as the atmosphere), the regions of high intensity within the speckle pattern are mostly unaffected by turbulence from both spectral and temporal point of views. In particular, the phase remains conserved, excepted for the linear group-velocity dispersion (GVD) which can be initially compensated by a negative chirp. This property, which is of prime importance for future remote coherent control experiments, have been demonstrated for narrow spectral bandwidths (30 nm i.e. 30 fs). Further studies are currently conducted for pulses with large spectral bandwidth and/or with central wavelength in spectral regions where the GVD fluctuations are not negligible such as in the UV. In the second section of this Chapter, the corpuscular nature of self-phase modulation has been highlighted. In particular, the study of correlations between the different spectral components allowed us to describe the supercontinuum generation as a four-wave mixing cascading. Moreover, by an adequate spectral filtering, the correlations have been used to significantly improve the intensity stability of the pulse after filamentation. This could be used in the future for the production of low noise white-light source.

Chapter 3 has been dedicated to the interaction between a filament and another ultrashort pulse. First, we have demonstrated that the high intensity within the filament core can significantly break the initial symmetry of the gas. Consequently, when a probe pulse co-propagates with the driving filament, it experiences a strong birefringence. We have demonstrated that the ability of the filament to carry high intensities over long distance can lead to the generation of an ultrafast half-wave plate, with a commutation time theoretically limited to the filament
duration. In the second part, we have studied, both experimentally and numerically, the interaction between two filaments of different color. The strong interaction driving the co-propagation leads to a new propagation regime, named "co-filamentation". In particular, both the two spectra and the two plasma channels generated by the individual filaments connect. Moreover, we have shown numerically that co-filamentation can lead to simultaneous cross-compression of the two pulses up to a factor 6, resulting in the generation of two intense 5-cycles pulses without any external temporal post-compression. This result could be useful for the generation of attosecond pulses.

Finally, in Chapter 4, we have described the first demonstration of the propagation of $20\ J$, $30\ TW$ laser through the atmosphere. The highly nonlinear propagation of the beam in the air gives rise to more than 400 self-guided filaments. This extremely powerful bundle of laser filaments generates a supercontinuum propagating up to the stratosphere, beyond 20 $km$. This constitutes the highest power "atmospheric white-light laser" to date.

The works conducted during this thesis could find direct applications for domains spanning from coherent (optimal) control in bio-molecules, to lightning triggering, from pollutants sensing, to attosecond pulse generation, and from noise reduction and ultrafast switching for remote optical communication, to phase-encrypted pulse transmission through the atmosphere.
Linear LIDAR theory

Standard LIDAR (Light Detection and Ranging) is based on the effect of optical scattering of light on atmospheric constituents. The Rayleigh-, Raman-, fluorescence, or Mie-scattering processes at a distance $z$ all return a small portion of an incident light back to the observer. Generally speaking, LIDAR is used to detect and identify trace substances in the atmosphere using a known laser source [173].

Elastic scattering form of the LIDAR equation

In the case of a pulsed LIDAR, the signal power $\Delta P(\lambda, z)$ received by the detector in the wavelength interval $[\lambda, \lambda + \Delta \lambda]$ from the element of altitude located in the interval $[z, z + \Delta z]$ is given by [174]:

$$\Delta P(\lambda, z) = \int_{\lambda}^{\lambda+\Delta\lambda} \int_{z}^{z+\Delta z} J(\lambda, z, \overrightarrow{r}) d\lambda dz p(\lambda, z, \overrightarrow{r}) dA(z, \overrightarrow{r})$$  \hspace{1cm} (5.1)

where

$J(\lambda, z, \overrightarrow{r})$ represents the laser-induced spectral radiance at wavelength $\lambda$, at position $\overrightarrow{r}$ in the target plane located at altitude $z$, per unit altitude interval;

$dA(z, \overrightarrow{r})$ represents the surface element of the target area at position $\overrightarrow{r}$ and altitude $z$;

$p(\lambda, z, \overrightarrow{r})$ represents the probability that radiation of wavelength $\lambda$ backscattered from position $\overrightarrow{r}$ at altitude $z$ hits the detector.

Many factors affect this probability. These include geometrical considerations, atmospheric attenuation, the receiver optics geometry, and spectral transmission characteristics. We can basically separate these influences and write:

$$p(\lambda, z, \overrightarrow{r}) = \frac{A_0}{z^2} T(\lambda, z) \xi(\lambda) \xi(z, \overrightarrow{r})$$  \hspace{1cm} (5.2)

where

$A_0/z^2$ represents the acceptance solid angle of the receiver optics ($A_0$ being the pupil object area of the receiver optics, which often corresponds to the primary mirror diameter);

$T(\lambda, z)$ represents the atmospheric transmission factor at wavelength $\lambda$ integrated up to the altitude $z$;
Linear LIDAR theory

$\xi(\lambda)$ represents the receiver’s spectral transmission factor and includes the influence of both the sensibility of the detector and a possible spectral filter placed in front of it;

$\xi(z, \vec{r})$, often referred to as the geometrical form factor, represents the probability of the radiation from position $\vec{r}$ in the target plane at altitude $z$ to reach the detector, based on geometrical considerations.

Since we consider the atmosphere as an elastic scattering medium, the target spectral radiance $J(\lambda, z, \vec{r})$ can be written as:

$$J(\lambda, z, \vec{r}) = \beta(\lambda, z, \vec{r})I(\lambda, z, \vec{r})$$  \hspace{1cm} (5.3)

where $I(\lambda, z, \vec{r})$ is the laser irradiance at position $\vec{r}$ and altitude $z$, and

$$\beta(\lambda, z, \vec{r}) = \sum_i N_i(z, \vec{r}) \frac{d\sigma(\lambda)}{d\Omega_i}$$  \hspace{1cm} (5.4)

is the volume backscattering coefficient, in which

$N_i(z, \vec{r})$ represents the number density of scatterer species $i$;

$\frac{d\sigma(\lambda)}{d\Omega_i}$ represents the differential scattering cross section of the species $i$ under irradiation with laser radiation at $\lambda$.

The total signal power $P(\lambda, t)$ received by the detector at the instant $t = 2z/c$, corresponding to the time taken for the laser pulse to propagate (at the velocity of light $c$) to altitude $z$ and the returned radiation to reach the LIDAR, can be expressed as:

$$P(\lambda, t) = A_0 \int_{z=0}^{z=ct/2} \frac{dz}{z^2} \int_{\Delta\lambda} d\lambda \int_A J(\lambda, z, \vec{r})p(\lambda, z, \vec{r})dA(z, \vec{r})$$  \hspace{1cm} (5.5)

Moreover, in our case, we will consider that the spectrum is flat and that all the variables are constant over the width $\Delta\lambda_0$ of the detection spectral range and assume that the probability $\xi(z, \vec{r})$ is essentially unity where the field of view of the receiver optics overlaps the laser beam and zero elsewhere (no vignetting). We will also assume that the transverse distribution of the pulse is "top-hat" over an area $A_L(z)$ at altitude $z$.

In this case,

$$P(\lambda, t) = A_0 \Delta\lambda_0 \xi(\lambda) \int_{z=0}^{z=ct/2} \beta(\lambda, z)T(z, \xi(z)I(z)A_L(z) \frac{dz}{z^2}$$  \hspace{1cm} (5.6)

An additional simplification is to approximate the temporal shape of the laser by a rectangle of duration $\tau$. This approximation is made for two main reasons: It makes the calculation easier and the real temporal shape of the pulse experiencing filamentation as a function of the altitude is obviously unknown. Then, the total power scattered laser power received at a time $t = 2z/c$ reads:

$$P(\lambda, t) = A_0 \Delta\lambda_0 \xi(\lambda) \beta(\lambda, z)T(z)\xi(z)I(z)A_L(z) \frac{cT_L}{2z^2}$$  \hspace{1cm} (5.7)
For a rectangular-shaped laser pulse of duration $\tau_L$,

$$I(z) = \frac{E_L(\lambda)T(\lambda, z)}{\tau_L A_L(z)} \quad (5.8)$$

where $E_L(\lambda)$ represents the energy of the laser pulse at the wavelength $\lambda$ ($E_L$ does not depend on $z$ because the laser pulse is considered to propagate linearly, i.e. the spectrum does not experience spectral broadening), $T(\lambda, z)$ represents the atmospheric transmission factor at the wavelength $\lambda$ to altitude $z$. This transmission factor follows the Beer-Lambert law, and consequently can be expressed as:

$$T(\lambda, z) = e^{-\int_0^z \kappa(\lambda, z) \, dz} \quad (5.9)$$

where $\kappa$ is the atmospheric extinction coefficient.

Finally, the radiative energy received by the detector during the interval $(t, t + \tau_d)$, where $\tau_d = 1/f$ is the integration time of the detection ($f$ is the frequency bandwidth of all the detection chain) and $t = 2z/c$ reads:

$$E(z) = E_L(\lambda)\Delta\lambda_0 \xi(\lambda)T^2(z)\xi(z)\frac{A_0}{z^2}\beta(\lambda, z)\frac{C_{td}}{2} \quad (5.10)$$

**Geometry of the receiver optics**

In this section, we will focus on the geometrical form factor $\xi(z)$ contained in the LIDAR equation derived previously (Eq. 5.10). We will assume that the pupil object is the primary mirror of the telescope and we neglect any obstruction from the secondary mirror. Moreover, we assume a flat laser distribution over the area of illumination. Under these circumstances, the geometrical probability factor $\xi(z, \overrightarrow{r})$, with $\overrightarrow{r}$ represented as $(r, \psi)$ is unity in the region of overlap between the laser beam and the filed of view of the receiver optics, and zero elsewhere. The geometrical form factor becomes:

$$\xi(z) = \frac{1}{\pi W^2(z)} \int_{r=0}^{r_T(z)} \int_{\psi=0}^{\psi=2\pi} \mathcal{H}(z, r, \psi) r \, dr \, d\psi \quad (5.11)$$

with

$$\mathcal{H} = \begin{cases} 
1 & \text{where the receiver-optics field of view and laser beam overlap} \\
0 & \text{elsewhere} 
\end{cases}$$

$W(z) = \sqrt{W_0 + \theta^2z^2}$ represents the beam radius, $\theta$ being the laser half-divergence and $W_0$ the laser output aperture radius. Under these conditions, we can write:

$$\xi(z) = \frac{A\{(r_T(z), W(z), d(z))\}}{\pi W^2(z)} \quad (5.12)$$

where

$A$ represents the area overlap function;
Figure 5.1: Geometry of a biaxial LIDAR, where the separation of the laser and telescope axes is \( d = d_0 - z\delta \) in the target plane. \( r_T \) is the radius of the circular filed of view and \( W \) is the radius of the circular region of laser illumination.

\[
r_t(z) = r_0 + \phi z \text{ is the radius of the receiver-optics field of view in the target plane; and}
\]

\[
d(z) = d_0 - z\delta \text{ is the separation of the telescope and laser axes in the target plane. Moreover,}
\]

\[
r_0 \text{ represents the radius of the object pupil of the telescope, } \phi \text{ represents the receiver-optics half field of view, } d_0 \text{ is the separation between the laser and telescope axes at } z = 0, \text{ and } \delta \text{ is the angle between the laser and telescope axes (see Fig. 5.1).}
\]

Three situations are possible:

1. The separation of the axes is too large so that there is no overlap between the receiver-optics field of view and the area of illumination: \( \mathcal{A} = 0 \) if \( d(z) > r_T(z) + W(z) \).

2. The separation of the axes is small enough that either the area of laser illumination lies totally within the receiver-optics field of view or vice-versa. This takes place when \( d(z) < |r_T(z) - W(z)| \) and \( \mathcal{A}(z) = \pi \times \min(r_T(z)^2, W(z)^2) \).

3. The separation of the axes lies between these two values:

\[
|r_T(z) - W(z)| < d(z) < r_T(z) + W(z) \tag{5.13}
\]

In this situation, the overlap function is:

\[
\mathcal{A} = W^2\Psi_W + r_T^2\Psi_z - r_Tdz\sin\Psi_z \tag{5.14}
\]

where

\[
\Psi_W = \arccos \left( \frac{d^2 + W^2 - r_T^2}{2Wd} \right) \tag{5.15}
\]
Figure 5.2: Variation of the overlap factor with the geometry of the receiver optics.
Figure 5.3: Example of a bad design of the receiver-optics geometry. The telescope becomes blind from about 100 m of altitude because of an under-estimation of the laser divergence.

\[ \Psi_z = \arccos \left( \frac{d^2 + r_T^2 - W^2}{2r_T d} \right) \]  

(5.16)

Figure 5.2 displays a few examples of the overlap factor as a function of the altitude for several geometries of the receiver optics. It clearly shows that a careful design of the detection setup is critical. In particular, as can be noticed in Fig. 5.3, a bad design can make the telescope completely blind at high altitude.

**Retrieval of the backscattering coefficient**

Both the air molecules of the air as well as the aerosols present in the atmosphere are responsible for light scattering. Because our experiments took place near the Atlantic ocean, where the humidity is high, we will neglect the Rayleigh scattering (induced by small molecules) and we will only consider the scattering induced by “big” water particles. The scattering efficiency and angular intensity diagram strongly depend on the aerosol shape and size. In most cases, there is no analytical solution to this problem. Under these circumstances, a classical approximation is to consider the particles as spherical and use the Mie scattering theory. The full description of this theory being beyond the scope of this section, we simply
remind that the Mie backscattering coefficient $\beta_{\text{Mie}}(\lambda)$ can be expressed as:

$$\beta_{\text{Mie}}(\lambda) = \int_0^\infty \pi r^2 n(r) Q_\pi(r, \lambda) dr$$  \hspace{1cm} (5.17)

where

$n(r)$ represents the distribution of the aerosol size (assumed spherical with radius $r$) within the atmosphere;

$Q$ represents the backscattering efficiency.

The backscattering efficiency $Q_\pi$ can be calculated from the Mie theory as:

$$Q_\pi(r, \lambda, n) = \frac{2}{r^2 k^2} \sum_{p=1}^{+\infty} (2p + 1)(|a_p|^2 + |b_p|^2)$$  \hspace{1cm} (5.18)

where

$n$ is the refractive index of the aerosol;

$$a_p = \frac{\psi_p(n.a)\psi'_p(a) - \psi_p(a)\psi'_p(na)}{\psi_p(n.a)\xi'_p(a) - \xi_p(a)\psi'_p(na)}$$  \hspace{1cm} (5.19)

$$b_p = \frac{\psi_p(n.a)\psi'_p(a) - n \cdot \psi_p(a)\psi'_p(na)}{\psi_p(n.a)\xi'_p(a) - n \cdot \xi_p(a)\psi'_p(na)}$$  \hspace{1cm} (5.20)

where $\psi_p(x) = \frac{i^{-p}}{2} \int_0^1 e^{ix\cos\theta} P_p(\theta) \sin \theta d\theta$, $\xi_p(x) \simeq (-i)^{p+1} e^{ix}$ as soon as $x \gg p^2$, and $P_p$ are the Legendre polynomials.

Figure 5.4 displays $Q_\pi(r, \lambda)$ calculated as described in Eq. 5.18 for a refractive index $n = 1.33$. This refractive index has been chosen because we assume that the main aerosol present during our experiments is water droplets.
Figure 5.4: Backscattering coefficient as a function of the incident wavelength and particle size. Note the oscillations corresponding to resonance frequencies of the aerosols.
Résumé

La filamentation est à l’origine de nombreuses transformations spatio-temporelles des impulsions laser ultrabrèves et de leur milieu de propagation. Le principal travail de cette thèse a été d’approfondir la compréhension de ce type très particulier de propagation. Ainsi, des simulations numériques, reproduisant la propagation des filaments dans les gaz, et plus particulièrement dans l’Argon, ont été entièrement développées. Ces simulations numériques, décrites dans le Chapitre 1, nous permettent de mieux apprêhender la physique sous-jacente la filamentation.

Expérimentalement, nous avons démontré que l’élargissement du spectre se fait de manière statistiquement ordonnée (les composantes spectrales générées sont fortement corrélées), ce qui a permis de mieux appréhender la dynamique de la génération du supercontinuum. En particulier, le lien entre automodulation de phase, vision classique de l’élargissement spectral, et mélange quatre ondes. Du fait des hautes intensités rgnant durant la propagation ($\approx 50 \, TW.cm^{-2}$), les filaments ont une forte influence sur la propagation d’une autre impulsion. De ce fait, nous avons étudié la copropagation de deux filaments et observé la concaténation des deux canaux plasma et le fort élargissement des spectres dû aux interactions croisées des impulsions. Parallèlement, nous avons montré que les filaments induisent une forte biréfringence, conduisant à la génération d’une lame demi-onde, pouvant théoriquement commuter la polarisation d’une sonde en quelques centaines d’attosecondes. Da’utre part, nous avons tudié l’influence de la turbulence sur la propagation d’impulsion ultracourte. Ainsi, nous avons pu démontré, à la fois expérimentalement et théoriquement, que l’action de la turbulence sur le profil temporel de l’impulsion peut être compensé activement en même temps que les tavelures induites sur le profil spatial.

Enfin, nous avons étudié la propagation d’un faisceau laser de 20 $J$, 32 $TW$ dans l’atmosphère, conduisant à la génération de plus de 400 filaments et d’un continuum de lumière blanche détectable jusque dans la stratosphère.

Tous ces travaux trouvent des applications dans de nombreux domaines, s’étendant du contrôle cohérent de bio-molécules à la génération d’impulsions attosecondes. De plus, les travaux sur la commutation ultrarapide et sur la réduction du bruit optique pourraient servir en télécommunication.
List of publications


Béjot P et al., 32 TW atmospheric white-light laser, APPL. PHYS. LETT. 90, 15 151106 (2007)


Bibliography


