Laser Filamentation - Beyond Self-focusing and Plasma Defocusing

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LASER FILAMENTATION – BEYOND SELF-FOCUSBING
AND PLASMA DEFOCUSING

by

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Diplôme d’Ingénieur, l’Institut d’Optique Théorique et Appliquée, 2004
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A dissertation submitted in partial fulfillment of the requirements
for the degree of Doctor of Philosophy
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Major Professor: Martin C. Richardson
ABSTRACT

Laser filamentation is a highly complex and dynamic nonlinear process that is sensitive to many physical parameters. The basic properties that define a filament consist of (i) a narrow, high intensity core that persists for distances much greater than the Rayleigh distance, (ii) a low density plasma channel existing within the filament core, and (iii) a supercontinuum generated over the course of filamentation. However, there remain many questions pertaining to how these basic properties are affected by changes in the conditions in which the filaments are formed; that is the premise of the work presented in this dissertation.

To examine the effects of anomalous dispersion and of different multi-photon ionization regimes, filaments were formed in solids with different laser wavelengths. The results provided a better understanding of supercontinuum generation in the anomalous dispersion regime, and of how multi-photon ionization can affect the formation of filaments.

Three different experiments were carried out on filamentation in air. The first was an investigation into the effects of geometrical focusing. A simplified theoretical model was derived to determine the transition of filamentation in the linear-focusing and nonlinear-focusing regimes. The second examined the effects of polarization on supercontinuum generation, where a polarization-dependent anomalous spectral broadening phenomenon due to molecular effects was identified. The third involved the characterization of filaments
in the ultraviolet. The combination of physical mechanisms responsible for filamentation in
the ultraviolet was found to be different from that in the near infrared.
ACKNOWLEDGMENTS

I would like to acknowledge the guidance and support of my advisor Dr Martin Richardson, who has given me this opportunity to delve into the amazing field of laser filamentation, and to be part of the LPL (Laser Plasma Laboratory) family. It has been a privilege to work with the people of LPL, and to be exposed to the diverse research that is conducted in the laboratory.

I would also like to thank the rest of my committee, Dr Demetrios Christodoullides, Dr Zenghu Chang and Dr Xi-Cheng Zhang, for their time and their insights.

Dr Matthieu Baudelet and Dr Magali Durand have been great friends and mentors. Despite their busy schedules, I have received countless nuggets of wisdom from them, be they about the physical sciences, the life of a researcher, or the French psyche. Dr Matthew Weidman was a wonderful lab mate to whom I am grateful for helping to familiarizing me with the laboratory. Together with Dr Mark Ramme, our many discussions over coffee have been the occasional source of little “ah-ha!” moments that have helped my research along.

Before working with the laser, we first had to build it (or more accurately, take it apart, and rebuild it). For this, I am grateful for the guidance of Dr Lawrence Shah, and the many hours of hard work by Benjamin Webb, Joshua Bradford and Nathan Bodnar. I hope that the results produced over the past year by the laser (a significant portion of this dissertation) shows that the many late nights and weekends have not been wasted.
My time in LPL and CREOL would certainly not have been as enriching without the support of friends and colleagues. It would be impossible to list them all, but special mention goes to some other members of the filamentation team who have been with me at various points of the journey: Dr Michael Chini, Dr Nicholas Barbieri, Erik McKee and Cheonha Jeon. The CREOL administrative staff also have my gratitude for helping to take care of the multitude of things that could have so easily distracted me from research work.

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I would also like to thank DSO National Laboratories in Singapore for providing the opportunity to pursue a PhD degree. Dr Desmond Lim, Dr Vincent Wong and Dr Vincent Leong have been nothing but encouraging of my endeavor.

Last but not least, my parents have always been an inspiration to me, and their shining example has been a driving force behind my achievements. And the support that mattered the most: my wife Sy Wei who agreed to walk this long road with me. This journey would not have been possible without her.
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<td>angle of incidence</td>
</tr>
<tr>
<td>AR</td>
<td>anti-reflection</td>
</tr>
<tr>
<td>BBO</td>
<td>beta-barium borate</td>
</tr>
<tr>
<td>CCD</td>
<td>charge-coupled device</td>
</tr>
<tr>
<td>CP</td>
<td>circular polarization</td>
</tr>
<tr>
<td>CPA</td>
<td>chirped pulse amplification</td>
</tr>
<tr>
<td>DSLR</td>
<td>digital single lens reflex</td>
</tr>
<tr>
<td>ETWM</td>
<td>effective three-wave mixing</td>
</tr>
<tr>
<td>FIBS</td>
<td>filament-induced breakdown spectroscopy</td>
</tr>
<tr>
<td>FWHM</td>
<td>full-width at half-maximum</td>
</tr>
<tr>
<td>GaAs</td>
<td>gallium arsenide</td>
</tr>
<tr>
<td>GVD</td>
<td>group velocity dispersion</td>
</tr>
<tr>
<td>iCCD</td>
<td>intensified charge-coupled device</td>
</tr>
<tr>
<td>IR</td>
<td>infrared</td>
</tr>
<tr>
<td>KSF</td>
<td>Kerr self-focusing</td>
</tr>
<tr>
<td>LIBS</td>
<td>laser-induced breakdown spectroscopy</td>
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<tr>
<td>LIDAR</td>
<td>light detection and ranging</td>
</tr>
<tr>
<td>LP</td>
<td>linear polarization</td>
</tr>
<tr>
<td>MPA</td>
<td>multi-photon absorption</td>
</tr>
<tr>
<td>MPI</td>
<td>multi-photon ionization</td>
</tr>
<tr>
<td>MTFL</td>
<td>Multi-Terawatt Femtosecond/Filamentation Laser</td>
</tr>
<tr>
<td>Abbreviation</td>
<td>Full Form</td>
</tr>
<tr>
<td>--------------</td>
<td>-----------</td>
</tr>
<tr>
<td>NA</td>
<td>numerical aperture</td>
</tr>
<tr>
<td>ND</td>
<td>neutral density</td>
</tr>
<tr>
<td>NIR</td>
<td>near-infrared</td>
</tr>
<tr>
<td>NLSE</td>
<td>nonlinear Schrödinger equation</td>
</tr>
<tr>
<td>OPA</td>
<td>optical parametric amplifier</td>
</tr>
<tr>
<td>QWP</td>
<td>quarter-waveplate</td>
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<td>second harmonic generation</td>
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<td>self-phase modulation</td>
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<tr>
<td>ZnSe</td>
<td>zinc selenide</td>
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CHAPTER 1 - INTRODUCTION

Figure 1: Picture of a laser filament in the laboratory, taken with a standard DSLR camera.

Laser filamentation has gained significant attention with the development and availability of high-power ultrashort pulse lasers. In the regime where laser filamentation takes place, the beam self-focuses as it propagates, eventually evolving into a very fine filament of high optical intensity. The filament is effectively non-diffracting and can maintain its dimensions for distances much longer than the Rayleigh length of an equivalent Gaussian beam.

Chiao et al. first proposed the possibility of a self-trapped beam of light in 1964 [1], and the same team demonstrated its existence in a carbon disulfide cell in 1966 [2]. The formation of laser filaments in liquids and in solids (for example, in [3] and [4] respectively) were subsequently reported and investigated. However, interest in laser filamentation remained
relatively limited until 1995, when Braun et al. first demonstrated the formation of a laser filament in air [5]. Since then, the field has developed rapidly, and its progress has been well documented in several recent publications such as references [6–10].

The objective of this dissertation is to investigate the outcome when deviating from the standard filamentation conditions – linearly polarized 800 nm pulses filamenting in air – and to understand how the filamentation process is modified by these changes. To facilitate this investigation, two tools, described in Chapter 2, have been developed: an imaging system for recording the beam profile of the filament and a simulation code to study the evolution of a filamenting pulse. Chapter 3 describes two experiments that were conducted in solids to examine the effects of dispersion regimes and multi-photon absorption regimes. Chapter 4 discusses filamentation in air, first in different focusing conditions, then with different polarization, and finally at a different wavelength. The findings in this dissertation will contribute to our overall understanding of filamentation science, and facilitate the deployment of laser filaments for potential applications. The remainder of Chapter 1 will be devoted to summarizing the basics of filamentation theory and highlighting various filament properties that will be discussed in greater detail in the following chapters.

1.1 - Filamentation Theory

Laser filamentation is a highly complex and dynamic physical phenomenon that results from the nonlinear interaction between an intense optical field and the medium through which it propagates. The medium in question can be of any physical state (solid, liquid or
gaseous), as long as it is transparent to the wavelength of the propagating laser pulse. For the sake of clarity and conciseness, this section will primarily describe laser filamentation in air. The wavelength of interest will be assumed to be $\lambda_0 = 800$ nm, which is representative of mode-locked Ti:sapphire laser systems frequently used for the creation of filaments. Ultrafast sources are necessary as (i) high peak power is an essential condition for self-focusing to occur and (ii) the pulses need to be short enough such that avalanche ionization does not dominate and lead to high values of plasma density that will upset the dynamic balance in the physical processes responsible for filamentation. Filamentation in condense media follows the same principles, but may differ in certain details, such as the arresting mechanism that counters self-focusing.

1.1.1 - Kerr Effect and Self-focusing

The optical Kerr effect is a $\chi^{(3)}$ nonlinear effect, referring to the change in a material’s refractive index in response to the optical field propagating in it. It is described by the nonlinear refractive index $n_2$, such that the effective index of the material is given by

$$n(r, t) = n_0 + n_2 I(r, t)$$ (1)

where $n_0$ is the linear refractive index of the material and $I(r, t)$ is the intensity of the field at a specific point in space and time. As a Gaussian beam propagates through air, the center of the beam therefore experiences a zone of higher index and the resultant wavefront deformation causes the beam to converge, akin to the effect of a positive lens.
Self-focusing, a concept first described by Kelley [11], occurs when the Kerr lensing effect is significant enough to overcome beam divergence due to diffraction. If this condition is met, the Kerr lensing effect is self-reinforcing as the beam propagates, and the beam shrinks rapidly in size. If an equilibrium could be achieved between self-focusing and diffraction, self-trapping of the beam occurs [1], and the beam propagates without any change in size, effectively behaving like a spatial soliton. However, this equilibrium is unstable [12] and cannot be used on its own to explain the formation of filaments. Nonetheless, self-focusing is the precursor to filamentation, and the Kerr effect is a key process in maintaining the integrity of the filament as it propagates.

For self-focusing to occur, the optical power contained in the beam has to exceed the critical value

$$P_{cr} = \alpha \frac{\lambda_0^2}{4\pi n_0 n_2}$$

(2)

$\alpha$ is a constant that depends on the transverse beam profile, and is typically on the order of 1.9 [13, 14]. In air $n_2 \approx 3 \times 10^{-19}$ cm$^2$/W [15], and critical powers of 3 – 10 GW are
typically quoted in the literature. For a Gaussian beam, once self-focusing sets in, the beam reaches a focus at the distance

\[
z_{sf} \approx \frac{0.367 ka^2}{\sqrt{\left(\frac{P}{P_{cr}} - 0.852\right)^2 - 0.0219}}
\]

where \( k \) is the wavenumber and \( a \) is the \( 1/e^2 \) intensity beam radius. The distance \( z_{sf} \) is measured from the beam waist [16]. Depending on the beam size and power, \( z_{sf} \) may be too long, especially in a laboratory setting. It is a common practice to focus the beam using optics of focal length \( f \), whereby the modified self-focusing distance \( z'_{sf} \) becomes [17]

\[
\frac{1}{z'_{sf}} = \frac{1}{f} + \frac{1}{z_{sf}}
\]

1.1.2 - Photo-ionization and Plasma Defocusing

When the beam size is greatly reduced from self-focusing, the electric field in the beam becomes intense enough to ionize the air through which it propagates. Two photo-ionization processes take place: multi-photon ionization (MPI) and tunneling ionization. MPI occurs when the electron simultaneously absorbs multiple photons, providing it with enough energy to overcome the ionization potential \( U_i \). In air, oxygen is the first element to undergo MPI with \( U_i \approx 12 \) eV. The photons in a \( \lambda_0 = 800 \) nm beam each have energy \( \hbar \omega \approx 1.55 \) eV, therefore 8 photons are necessary to overcome \( U_i \) and the MPI rate will scale as \( I^8 \). Nitrogen, with ionization energy of 15.6 eV, will also be ionized in a filament, but its contribution is negligible and can be omitted for the sake of simplicity without
affecting the validity of the discussion and simulation results in the following chapters [18]. Tunneling ionization describes the escape of an electron from the Coulomb potential of the nucleus, which is highly perturbed by the external field. Both ionization processes require very high field/intensity, and therefore only occur when the beam is focused, either by external focusing or self-focusing.

Figure 3: (a) MPI occurs when the electron absorbs multiple photons, providing it with sufficient energy to overcome the ionization potential. (b) Tunneling ionization occurs when the external electric field is strong enough to distort the Coulomb potential around the atom, allowing the electron to escape. Source: [6]

By calculating the Keldysh parameter $\gamma$, the regime of photo-ionization that is dominant in laser filamentation can be determined. $\gamma$ is given by

$$\gamma = \frac{\omega}{e} \sqrt{\frac{m_e U/n_0 c e_0}{I}}$$

where $\omega$ is the laser angular frequency, $m_e$ and $e$ the mass and charge of an electron respectively. $c$ is the speed of light in vacuum and $e_0$ is the permittivity of free space. MPI dominates if $\gamma \gg 1$, and tunneling ionization dominates if $\gamma \ll 1$. For the case of a $\lambda_0 = 800$ nm beam in air and intensities typical of a filament, $\gamma \approx 1.8$. In such a case, the Keldysh formulation should be used to determine the ionization rate [19–21]. Nonetheless,
approximation of the ionization rate by MPI has been shown to provide satisfactory agreement with experimental observations [20, 22], and this approach will be adopted for this dissertation.

The evolution of the electron density in the filament can be described by

\[ \frac{\partial \rho}{\partial t} = \sigma_K I^K (\rho_{nt} - \rho) + \frac{\sigma}{U_i} \rho I \]  

(6)

The first term on the right hand side represents \( K \)-photon MPI, and the second represents avalanche ionization. \( \sigma_K \) is the \( K \)-photon ionization cross section \( \sigma_8 = 2.81 \times 10^{-96} \text{ W}^{-8} \text{ cm}^{16} \text{s}^{-1} \) for oxygen, \( \rho_{nt} \) the density of neutrals in the medium \( \rho_{nt} = 0.54 \times 10^{19} \text{ cm}^{-3} \) for 20\% composition of oxygen in air and \( \sigma = \frac{k_0}{n_0 \rho_c} \frac{\omega_0 \tau_c}{(1 + \omega_0^2 \tau_c^2)} \approx 5.6 \times 10^{-20} \text{ cm}^{2} \) is the cross section for inverse Bremsstrahlung. \( \tau_c \approx 350 \text{ fs} \) is the electron collision time and \( \rho_c = \epsilon_0 m_e \left( \frac{2 \pi e}{\epsilon_0 h} \right)^2 \approx 1.7 \times 10^{21} \text{ cm}^{-3} \) denotes the critical plasma density above which the plasma becomes opaque. \( \epsilon_0 \) is the permittivity of vacuum, \( m_e \) and \( e \) the mass and charge of an electron, and \( c \) the speed of light in vacuum. Since \( \omega_0 \tau_c \gg 1 \), the approximation \( \sigma \omega_0 \tau_c \approx \frac{k_0}{n_0 \rho_c} \) can be made.

The presence of free electrons reduces the refractive index of the medium. Based on the Drude model, the change in index is given by

\[ n(\mathbf{r}, t) = n_0 - \frac{\rho(\mathbf{r}, t)}{2 \rho_c} \]  

(7)
where $\rho(r, t)$ denotes the density of free electrons at a specific point in time and space. The index change due to the plasma created works in opposition to the Kerr effect, countering the effects of Kerr self-focusing as illustrated in Figure 2.

1.1.3 - Dynamic Spatial Replenishment and the Nonlinear Schrödinger Equation
Mlejnek et al. first proposed the dynamic spatial replenishment model, which is now the most widely-accepted description of the filamentation process [23, 24]. They described filamentation as a dynamic exchange of energy between the filament and an energy reservoir surrounding it. The beam can be treated as a series of infinitesimally fine slices in time. The central time slice of the pulse has the highest power and therefore reaches a focus first, ionizes the air and gets defocused into the surrounding energy reservoir. The leading slices of the pulse do the same at progressively longer distances. The trailing slices of the pulse self-focus and encounter the plasma generated by the central and leading slices, which causes them to diverge in the form of transverse rings [25–27]. With further propagation, the focusing-defocusing cycle will repeat for the portions of the pulse that still carries sufficient power. The leading slices eventually decays due to energy lost to MPI, while the trailing slices refocus and feed energy back into the filament from the reservoir, sustaining it for longer distances. This focusing-defocusing cycle can occur multiple times and manifests itself as hot-spots along the filament.
By using the slowly varying envelop assumption and taking a reference frame moving along with the pulse at its group velocity, the evolution of the filament described by dynamic spatial replenishment can be derived from the nonlinear Schrödinger equation (NLSE)

\[
\frac{\partial \varepsilon}{\partial z} = \frac{i}{2k} \nabla^2_\perp \varepsilon + i k_0 n_2 |\varepsilon|^2 \varepsilon - i \frac{k_0 \rho}{2 n_0 \rho_c} \varepsilon
\]  

(8)

On the right hand side, the terms describe the effects of diffraction, Kerr self-focusing and plasma defocusing. Here, \(\nabla^2_\perp = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\) is the transverse Laplacian operator and \(k\) and \(k_0\) are the wavenumber in the medium and in vacuum respectively. \(\varepsilon\) is defined such that \(I = |\varepsilon|^2 = \frac{1}{2} n_0 \varepsilon_0 c |E|^2\).

Equation (8) is sufficient to describe the basic premises of the dynamic spatial replenishment model. However, to obtain simulation results that accurately reflect the measurements taken in the laboratory, other effects may need to be taken into account. An expanded version of the NLSE has been used by several authors [6, 23, 24, 28–31].
The terms on the right hand side now include the effects of diffraction, group velocity dispersion (GVD) \( k'' \), instantaneous Kerr effect, delayed Raman-Kerr effect, plasma absorption and defocusing, and MPI losses. \( R(t) = \frac{\nu - \omega_R^2}{\omega_R^2} \exp(-\Gamma t) \sin \omega_R t \) is a function that describes the molecular response of the medium, with \( \Gamma^{-1} \) being the typical molecular response time and \( \omega_R \) the molecular rotational frequency. In typical atmospheric conditions, \( \Gamma = 1/70 \) fs and \( \omega_R = 16 \times 10^{12} \) s\(^{-1} \). The factor \( f \) repartitions the Kerr self-focusing effect to the instantaneous electronic component and the delayed molecular Raman component. \( f = 0.5 \) has been accepted to provide a good fit in typical conditions. \( \beta_K = K \hbar \omega_0 \rho_{nt} \sigma_K \) is the MPI coefficient. Equation (9), together with Equation (6) form the basis for simulation work that is discussed in Chapter 2.2.

1.2 - Properties of Filaments

The laser filament’s non-diffracting nature may lead a new-comer to the field to compare it to other non-diffracting optical beams such as Bessel and Airy beams. Some authors have also referred to the filamenting pulse as a “light-bullet”, which may lead the reader to think of it as a “simple” spatial soliton. In reality, the filament possesses unique properties which set it apart from these other propagation regimes. It is self-starting and self-sustaining,
without needing any kind of beam engineering; It has soliton-like properties even though it is constantly evolving and does not have a stable solution; Its interaction with the propagation medium is of a much higher complexity thanks to its high intensity and the creation of the plasma channel.

Before the use of laser filaments can be integrated into more complex systems with real-world applications, their properties have to be properly determined and understood. Characterization of the filament is important both as a means to verify the theories relating to its formation and existence, and to evaluate its value in potential applications. Direct measurement of certain filament parameters can be difficult due to the high intensity of the filament, which will damage any optics or detection devices placed in its path. Indirect measurement techniques are often employed, and the relevant filament characteristic inferred. Results published by various research groups to date have often not been in total agreement. This is due to differences in the techniques used and in experimental conditions as well as to the lack of standardized definition of certain parameters. To have a good idea of the typical numbers obtained, the reader can refer to the summary of some filament characteristics measured between 1995 and 2002 published by Khan et al. [32].
Table 1: Compilation of filament characteristics, extracted from Khan et al. [32]

<table>
<thead>
<tr>
<th>Laser and medium</th>
<th>Braun et al. [5]</th>
<th>Nibbering et al. [33]</th>
<th>Brodeur, Kosareva et al. [34, 35]</th>
<th>Lange et al. [36]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength (nm)</td>
<td>775</td>
<td>800</td>
<td>800</td>
<td>800</td>
</tr>
<tr>
<td>Pulse length (fs)</td>
<td>200</td>
<td>150</td>
<td>220</td>
<td>120</td>
</tr>
<tr>
<td>Pulse energy (mJ)</td>
<td>20</td>
<td>30</td>
<td>10</td>
<td>5</td>
</tr>
<tr>
<td>Peak power (GW)</td>
<td>80</td>
<td>160</td>
<td>36</td>
<td>33</td>
</tr>
<tr>
<td>Critical power (GW)</td>
<td>1.7</td>
<td>3.4</td>
<td>6.1</td>
<td>3.6</td>
</tr>
<tr>
<td>n$_2$ (cm$^2$W$^{-1}$)</td>
<td>5.6 x 10$^{-19}$</td>
<td>3 x 10$^{-19}$</td>
<td>1.57 x 10$^{-19}$</td>
<td>2.8 x 10$^{-19}$</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Filaments</th>
<th>No. of filaments</th>
<th>Filament size (μm)</th>
<th>Propagation distance (m)</th>
<th>Energy content/filament (mJ)</th>
<th>Electron density (cm$^{-3}$)</th>
<th>Filament intensity (W cm$^{-2}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>3</td>
<td>80-100</td>
<td>50</td>
<td>0.75</td>
<td>10$^{16}$</td>
<td>7 x 10$^{13}$</td>
</tr>
<tr>
<td></td>
<td>≥2</td>
<td>250</td>
<td>111</td>
<td>0.7-1</td>
<td></td>
<td>10$^{14}$</td>
</tr>
<tr>
<td></td>
<td>1</td>
<td>190</td>
<td>100</td>
<td>0.34</td>
<td></td>
<td>10$^{14}$</td>
</tr>
<tr>
<td></td>
<td>Several</td>
<td></td>
<td></td>
<td>0.5</td>
<td></td>
<td>10$^{14}$</td>
</tr>
</tbody>
</table>

Table 1 only summarizes a very limited subset of the filaments’ properties, and only those that are relatively easy to quantify. More elaborations of some of these properties, as well as others not represented in the table, are given in the following paragraphs.

**Filament dimensions.** The size of a filament typically refers to just the diameter of the bright central core, sans the energy reservoir surrounding it. It can be estimated by measuring ablation or burn spots caused by the filaments, or by terminating the filamentation process (either by direct attenuation of the beam [37] or by a change in the propagation medium [38, 39]) before imaging the beam. The filament’s dimensions are also frequently inferred from the size of its plasma channel, which is easily visible due to fluorescence emission from the excited N$_2$ and N$_2^+$. 
**Intensity clamping.** As the self-focusing and defocusing (MPI) effects are both dependent on the intensity of the beam, it stands to reason that the intensity in a filament needs to stay in a range where the dynamic equilibrium between the Kerr focusing and plasma defocusing effects can be maintained. Since the ionization rate has a $I^K$ dependence, fluctuations in the intensity stay relatively small and the intensity is “clamped” for the length of the filament. This phenomenon of intensity clamping in a filament was described in the first experiments by Braun *et al.* [5], elaborated upon by Kasparian *et al.* [40] and shown experimentally by Becker *et al.* [41]. Since the temporal profile of the filament can evolve rapidly, accurate information on the intensity can only be deduced by observing a well calibrated intensity-dependent processes [42, 43]. The critical intensity from both simulations and experiments in nitrogen or air lies in the range of $10^{13} - 10^{14}$ W/cm².

**Plasma channel.** The plasma channel is an integral part of the laser filament, not only supporting its existence but also providing it with the potential for some unique applications such as discharge guiding [44–47], microwave guiding [48–50] and inducing condensation [51–53]. Due to the longer plasma recombination time compared to the pulse duration, the plasma channel can persist for up to several nanoseconds after the passage of the laser pulse, although its density decays rapidly after its formation. The plasma density in the channel is typically in the range of $10^{16} - 10^{17}$ cm$^{-3}$, but can be significantly different depending on the focusing conditions of the beam [54]. Depending on the application, it is also possible to increase the plasma’s density and lifetime using an igniter-heater scheme [55, 56].
Supercontinuum generation and conical emission. A highly prominent by-product of the filamentation process is the white-light supercontinuum [33, 57–60]. As the filament propagates, its spectrum broadens and an image of its cross-section will reveal a white central zone surrounded by rainbow-colored rings, often referred to as conical emission. The generation of white light is largely due to self-phase modulation (SPM) – the temporal manifestation of the Kerr effect. New frequencies are generated as the field in the pulse is red-shifted at the rising edge and blue-shifted at the falling edge. The tight spatial confinement of energy along the filament provides a long interaction length for significant spectral broadening to take place. In addition, self-focusing of the beam, leading to self-steepening of the pulse and ionization of the medium, amplifies the spectral broadening compared to SPM acting alone [9, 60–62]. The generation of supercontinuum makes it possible to extract shorter pulses from a filament [22, 63–65], and allows filaments to be used in spectroscopy and sensing applications [66].

1.3 - Applications of Filamentation

As a new propagation regime with vastly different properties from previously known ones, filamentation has not only expanded on current applications of ultrafast lasers, it has also paved the way for various novel applications. Having an intense channel of light that can propagate without suffering from the effects of diffraction is highly attractive for many nonlinear or remote sensing applications. The creation of long plasma channels and a white-light continuum also play crucial roles in potential novel applications. As it would be
impractical to present all the possible applications of filamentation, this section discusses some of the applications that are currently actively pursued by various research groups.

1.3.1 - Guiding Applications

The plasma density in the filament's plasma channel is known to be several orders of magnitude above the required free-electron density for initiating electrical discharges in the atmosphere [67]. It has been demonstrated that filaments bridging the gap between highly charged electrodes will trigger and guide electrical discharges between them [68]. There have been investigations into the feasibility of deploying laser filaments as lightning protection devices [32, 67, 69]. However, challenges remain in maintaining the guiding channel for a sufficient length in both space and time to achieve cloud-to-earth guiding. On a smaller scale, electrical discharge guiding with filaments can be used to generate ad-hoc antennas [46] and to trigger spark gaps [44].

Other than guiding electrical discharges, which are basically large bursts of current, filaments can also be used to guide electromagnetic radiation, typically in the radio-frequency range [70]. In its simplest form, a single filament or a pair of filaments can behave like conducting wires, and are capable of carrying radio-frequency signals along their length [71, 72]. Alternatively, arrays of filaments can form waveguiding structures. Various structures, based on different waveguiding principles, can be used. Due to the lower refractive index in the plasma channels, a ring of filaments will form a waveguide with the unionized air in the middle acting as the waveguide core [73]. If the ring of
filament is sufficiently dense, it can also act as a conductive barrier that traps and guides RF waves, which have frequencies lower than the filaments’ plasma frequency [48, 49]. A third possibility treats a regular array of filament plasma channels as a virtual hyperbolic metamaterial, where the anisotropic effective permittivity of the medium causes the RF radiation to be focused, guided or steered in the desired direction [50].

The guiding of optical wavelengths by filament arrays is has also been shown to be possible via index guiding. The index change exploited need not solely be that of the plasma channel [74], but can also be from the change in air density and temperature after the passage of the filamenting pulse [75].

1.3.2 - Sensing Applications
Remote sensing applications were some of the first to be explored after the initial observation of filamentation in air. The ability to maintain high intensities and fluences over long distances, as well as the generation of a supercontinuum that can span multiple octaves, are properties that make filaments the ideal candidate for various remote sensing techniques [76, 77]. Some notable examples of remote sensing applications are filament-induced breakdown spectroscopy (FIBS), white-light LIDAR, and terahertz generation and detection.

FIBS is a natural extension of the laser-induced breakdown spectroscopy (LIBS) technique [78], where a filament is used as the source in place of the typical focused nanosecond
pulse. Filamentation makes it possible to deliver the intensities necessary to generate LIBS signals to distances that would be impractical or impossible with conventional optics, allowing LIBS analysis to be performed at stand-off distances [79–82]. FIBS has been demonstrated at distances up to ~100 m and on solid, aerosol and gaseous targets [83, 84].

The supercontinuum generated by a filament can serve as a remote white-light source for absorption LIDAR measurements. The backscattered supercontinuum light is selectively absorbed by the media it traverses, and the absorption spectrum of the media can be deduced. The absorption lines of oxygen and water vapor in the atmosphere have been identified in a demonstration of such a filament-based LIDAR technique [66, 85], and the possibility of detecting trace gases has also been investigated [86].

The emission of energetic terahertz pulses from filaments [87, 88], especially ones generated by both the fundamental wavelength and its second harmonic [89, 90], means that they can be deployed as remote terahertz sources. This is especially important as terahertz radiation propagates poorly in the atmosphere [91], making remote terahertz sensing impossible by traditional methods. Air plasma-based techniques to detect terahertz waves are also being developed [92, 93], with the hope of eventually applying them with filaments and attaining a complete remote terahertz sensing system [94, 95].
1.3.3 - Other Unique Applications

Two other applications that are currently active areas of research are filament-induced condensation and the lasing effect in air.

When filaments propagated through cloud chambers, the aerosols in the chambers have been observed to coalesce and even form snow in the right conditions [51–53]. The plasma created in the filaments leads to the formation of larger molecules that serve as condensation nuclei. LIDAR measurements suggest that a similar phenomenon can be observed with filaments propagating in the atmosphere [51].

The presence of optical gain and amplified stimulated emission in a filament was first observed in 2003 [96]. In more recent years, various research groups have examined this effect [97–100], which is attributed to the emission linked to the first negative band ($B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+$) of $N_2^+$ and the second positive band ($C^3\Pi_u \rightarrow B^3\Pi_g$) of $N_2$. Amplified emissions in the filaments at various spectral lines in the violet and ultraviolet wavelengths have been reported, both in the forward and backward directions [101].
CHAPTER 2 - VISUALIZING FILAMENTS

As a laser filament is maintained by a highly dynamic balance of linear and nonlinear processes, the profile of a filamenting pulse is constantly evolving. To fully appreciate the physics involved and how different processes manifest during filamentation, it is essential to follow the evolution of the pulse. This can be done both numerically and experimentally, and both approaches will be tackled in this dissertation.

As outlined in Chapter 1.2, there are many physical properties of interest in a filamenting pulse. Many experiments of various complexities have already been designed to measure these properties, which include the fluence profile [37–39], peak intensity [43], temporal profile [39, 102, 103], and plasma density [104–107], among others. This dissertation has chosen to focus on the fluence profile, more simply referred to as the beam profile, as it most directly impacts other filamentation related research that our laboratory's filamentation team is involved in. It is possible to track all the properties of the filament through simulations, but computational time and resources impose limitations on the scenarios that can be effectively studied. For this dissertation, the goal is to obtain relatively quick and representative simulation results that can help in the qualitative prediction of experimental outcomes, without necessarily obtaining a high degree of precision.
2.1 - Filament Beam Profile Measurements

On the surface, measuring the beam profile of a filament appears to be the obvious approach for studying the spatial evolution of the filamenting beam. However, due to the high intensities in the filament, it is difficult to insert the necessary attenuators or beam samplers without causing damage to them. One effective method to obtain a beam profile of the filament at any point along its propagation axis is to change the propagation medium to one whose nonlinearity is much weaker. Examples of such a transition are from air to helium [38], air to vacuum [39], and water to air [108]. For measurements of filaments in air, such a transition usually requires the alignment of the filament through a small aperture on a gas chamber. The measurement cannot be easily translated longitudinally along the filament and is more suitable for measuring filaments assisted by medium-distance focusing (<1 m to a few meters) that have high pointing stability. Another method is to insert an attenuator or sampler in the beam in a way that ensures its integrity. The use of a grazing incidence reflection to sample a filament was first reported by Lange et al. [36]. The University of New Mexico group has used a suitably coated grazing angle reflector that sufficiently attenuates the transmitted beam to successfully image the filament directly [37, 109]. This method allows for the beam sampler to be easily translated along the filament, and ensures that the energy reservoir around the filament is not truncated by an aperture.

2.1.1 - Grazing Incidence Imaging System

The imaging system designed for this dissertation is similar in principle to that which was first reported by Lange et al. [36] – an uncoated fused silica wedge is placed at grazing
incidence in the filamenting beam, and the reflection is imaged onto a CCD camera. The angle of incidence (AOI) was chosen based on two principal considerations: the intensity of the filament has to be distributed over a large enough area on the wedge to avoid surface damage, and the reflectance has to be kept as low as possible. To find the range of AOI where surface damage would not occur, a filament was generated by focusing 5 mJ, 48 fs, 10 Hz pulses with a $f = 2$ m lens. Surface damage occurred when the AOI was smaller than 79° (with respect to the surface normal). To provide sufficient buffer from the damage zone and allow to small misalignments, the AOI on the wedge was chosen to be 83°. The intensity of the incident beam was reduced by 8.2x compared to normal incidence, and the reflectivity was 36% for P-polarized light.

As a single reflection could not provide enough attenuation for the beam to be safely imaged on a CCD, a total of five wedges were used – the first two wedges at 83° AOI, the next two at 65° (~1% reflectivity each), and the fifth wedge at 45° (~1% reflectivity). The intensity and fluence of the beam was therefore reduced by a factor of $10^7$, and it could be safely imaged onto a CCD camera with a lens. Additional ND filters could be inserted into the attenuated beam to further reduce the intensity when necessary. The CCD camera used (The Imaging Source DMK72BUC02) had a sensor size of 5.76 $\times$ 4.29 mm, 2.2 µm pixel size and 8-bit dynamic range. The imaging lens ($f = 100$ mm biconvex lens) imaged the plane intersecting the middle of the first wedge onto the camera CCD with 1x magnification.
2.1.2 - Filament Measurements at Various Distances

The imaging system was used to examine the profile of filaments generated with different focusing conditions and at different distances.

- Short distance: beam focused with a 2.15 m lens, 5.3 mJ 50 fs pulses
- Medium distance: beam focused with a 11 m lens, 5.5 mJ 50 fs pulses
- Long distance: collimated beam, 20 mJ ~500 fs negatively chirped pulses

The laser output is Gaussian in profile with a FWHM of 5.3 mm, and its spectrum is centered at 800 nm.
2.1.2.1 - Short Distance Measurements

Figure 6. Beam profiles of the filament taken at short distance with a 2.15 m focusing lens. The FWHM of the profiles in the x- and y-axes have been plotted together with the theoretical Gaussian profile for comparison.

For the short distance measurements, the profile of the filament evolved as expected. The focusing effect is linear up to a distance of $z = 1.6$ m, before the effect of Kerr focusing is observable and the beam converged more rapidly than a theoretical Gaussian beam. At $z \approx 2$ m, the beam was seen to collapse, and maintained a constant size of $\sim 85 \mu$m for a distance of more than 100 mm. Once past the linear focal point, the linear divergence of the beam dominated and the beam diverged at a similar rate as a linearly propagating beam. In the filamentation zone between $z = 2$ m and $z = 2.2$ m, the energy reservoir around the filament took on the profile of concentric rings, in agreement with what Chin et al. had observed [25–27].
2.1.2.2 - Medium Distance Measurements

Figure 7. Beam profile of the filament taken at medium distance with a 12 m focusing lens (blue squares and green circles). The dashed lines correspond to the respective mean FWHM values. For comparison, the FWHM of a theoretical Gaussian beam (red line) and the profile of a non-filamenting beam (purple crosses and black diamonds) focused by the same lens are plotted on the same graph. Note different scale on the profile images of the non-filamenting beam. Data collection was limited to a distance of 11 m by the dimensions of the laboratory.

In the medium distance measurements, the beam was also observed to focus more rapidly and attained a smaller beam size than a theoretical Gaussian beam. Beyond a distance of 9.5 m, the FWHM of the profile stabilized at ~200 μm. For comparison, the energy of the laser pulses was reduced from 5.5 mJ to 0.1 mJ, below the critical power necessary for
filamentation to occur, and the beam profile was measured. The non-filamenting beam had a significantly different profile, as it focused poorly and was noticeably astigmatic.

2.1.2.3 - Long Distance Measurements

![Beam profile taken at long distance. The beam was launched collimated and negatively chirped. The dashed lines correspond to the respective mean FWHM values. Inset is a representative beam profile taken at a distance of 36 m.](image)

The beam was negatively chirped to a duration of ~500 fs by translating the compressor grating, the pulse energy was turned up to 20 mJ, and the beam was sent to the propagation range in the CREOL chaseway. A filament began forming at a distance of ~10 m, verified by observing the ablation of a cardboard target. The stability of the filaments formed, both in size and position, was much poorer due to air turbulence and the lack of aid from focusing optics. Nonetheless, the beam was clearly less than 1 mm in diameter, and diverging very slowly over a distance of 40 m. The propagation was decidedly nonlinear; a theoretical Gaussian beam with 500 μm FWHM would maintain a similar level divergence over a Rayleigh distance of just 0.7 m. However, the beam size was significantly larger than that of a typical filament. The profile measured may therefore not be of a filament in the
traditional sense, but that of a similar high-intensity light channel generated by chirped pulses [110, 111].

2.1.3 - Application of Imaging System to Filamentation Research

A beam profiling system that can be easily displaced along the length of the filament is a useful tool in filamentation related research. The grazing incidence imaging system has been used to collect important data in two filamentation projects – ablation of solids by filaments and rotating helical filaments.

2.1.3.1 - Ablation of solids by filaments

As part of our laboratory’s efforts to investigate the interaction of filaments with solid targets, Weidman [112] made a detailed comparison of the measured filament fluence

![Comparison of the filament profile (top) with the ablation crater profile (bottom) on a GaAs sample. Source: [112]](image-url)
profile with the ablation crater profile on a GaAs target. The detailed filament profile, with fluence levels calibrated based on the ablation threshold of GaAs, allowed him to conclude on the impact of the filament energy reservoir on ablation. The difference in profile between a focus-assisted filament and one formed from a collimated beam has also been linked to variations in the ablation profiles in the two regimes.

2.1.3.2 - Rotating Helical Filaments

Figure 10. The rotating helical beams depicted in isometric view in the upper axes, and the beam profiles at various position of linearly propagating helical beams (top: simulation, middle: generated with 135 μJ) and filaments (bottom: generated with 11.5 mJ). Source: [113]
The combination of beam engineering with laser filamentation is of great interest to the laser filamentation community as it allows the filaments to be controlled and organized in potentially useful ways. In our laboratory, we have investigated filaments formed by rotating helical beams [70, 113, 114]. The grazing incidence imaging system verified the rotating helical structure of the beam, while ionization measurements confirmed that the beam created plasma channels. The profiles also revealed that the ionization process disrupted the helical structure, providing insight into the size limitations that structured filament arrays may face.

2.2 - Filament Simulation

The information provided by numerical simulations complement the experimental data collected in the laboratory. In a field such as laser filamentation, where many complex and nonlinear processes contribute to the overall observed phenomenon, it becomes even more valuable to have the appropriate numerical tools to help dissect the problem and analyze each contribution in a way that may not be possible in an experiment. The propagation of a filamenting laser pulse has already been modeled and solved via various numerical methods (for some examples, see references [31, 115–117]). It is not the scope of this dissertation to provide novel insight to numerical methods for laser filamentation, but rather to develop an in-house numerical tool for first-cut approximations to help predict, explain or verify experimental observations made by the laboratory.
2.2.1 - Solving the NLSE by the Split-step Method

The split-step method has been chosen as the technique for numerically solving the nonlinear Schrödinger equation (NLSE) as represented by Equation (9). To reduce the computation time and resources required, the beam is assumed to be rotationally symmetric, and the problem is solved in (2D+1) dimensions – the envelop and phase of the electric field is recorded in a 2 dimensional mesh whose axes represent the radial distance, \( r \), from the propagation axis and the local time, \( t \), with respect to the pulse (assumed to travel at a fixed group velocity). The evolution of the pulse is then tracked along the propagation axis.

When applying the split-step method on Equation (9), each propagation step is broken down into three sub-steps. The first sub-step treats only the diffraction term on the right-hand side. As the beam is radially symmetric, the diffraction term can be solved just in \( r \). To solve \( \frac{\partial \Phi}{\partial z} = \frac{i}{2k} \nabla_\perp^2 \varepsilon \), we can transpose the problem into the spatial frequency domain, whereby the operator \( \nabla_\perp^2 \) is transformed into a simple multiplication by \( -k_r^2 \), where \( k_r \) is the spatial frequency of the beam in the radial direction. Since the problem is treated in a single spatial dimension \( r \) and not \( x \) and \( y \), the Hankel transform is used instead of the Fourier transform. The first sub-step is therefore to solve

\[
\frac{\partial \tilde{\varepsilon}_r}{\partial z} = -i \frac{k_r^2}{2k} \tilde{\varepsilon}_r
\]

where \( \tilde{\varepsilon}_r \) is the Hankel transform of \( \varepsilon \) in the radial dimension. The analytical solution is
\[ \tilde{\varepsilon}_{r,n+1/3} = \tilde{\varepsilon}_{r,n} \exp \left( -i \frac{k_r^2}{2k} \Delta z \right) \]  

(11)

where \( \tilde{\varepsilon}_{r,n} \) and \( \tilde{\varepsilon}_{r,n+1/3} \) are the fields after the \( n \)th step and the \( n \)th step plus the first sub-step respectively, and \( \Delta z \) is the size of the step that is being taken. The second sub-step treats only the dispersion term. Similarly to the first sub-step, the problem can be solved in the spectral domain and we obtain

\[ \tilde{\varepsilon}_{t,n+2/3} = \tilde{\varepsilon}_{t,n+1/3} \exp \left( i \frac{k_\omega}{2} \Delta z \right) \]  

(12)

where the notation \( \tilde{\varepsilon}_t \) refers to the Fourier transform of \( \varepsilon \) in the time dimension. The third sub-step treats all the nonlinear terms by grouping them together:

\[ N = ik_0 n_2 (1 - f) |\varepsilon|^2 + ik_0 n_2 f \left[ \int_{-\infty}^{t} R(t - t') |\varepsilon(t')|^2 \, dt' \right] \]

\[ -\sigma \left( 1 + i \omega_0 \tau_c \right) \rho - \frac{\beta_k}{2} |\varepsilon|^{2k-2} \left( 1 - \frac{\rho}{\rho_{nt}} \right) \]

(13)

The nonlinear terms can therefore be applied by

\[ \varepsilon_{n+1} = \varepsilon_{n+2/3} \exp(N \Delta z) \]  

(14)

completing the calculations to move from the \( n \)th step to the \((n+1)\)th step.

Equations (11) and (12) describe linear propagation and therefore do not impose any limitations on the step size \( \Delta z \). Equation (14) describe self-action effects, and the resulting changes per step must be kept small to ensure that large errors are not accumulated over the propagation distance. For this purpose, the step size \( \Delta z \) is adaptive and changed over the course of the propagation to keep \( |N \Delta z| < 0.01 \). The resolution of the \((r, t)\) mesh also needs to be kept sufficiently fine to resolve the sharp spatial and temporal peaks that result
from filamentation. Since the beam does not collapse to a size smaller than \(\sim 100\ \mu m\) in a filament, a resolution of \(\sim 10\ \mu m\) is generally sufficient in \(r\). In the time axis, due to pulse splitting and shortening, a resolution of \(\sim 1\ fs\) is required.

At the beginning of each step, the plasma density \(\rho(r, t)\) is calculated based on the previous step’s intensity profile. Along the time axis at each radial position, \(\rho\) is determined by

\[
\Delta \rho(t_n) = \sigma_K \frac{I_K(t_n) + I_K(t_{n-1})}{2} (\rho_{nt} - \rho(t_{n-1})) \\
+ \frac{\sigma}{U_I} \rho(t_{n-1}) \frac{I(t_n) + I(t_{n-1})}{2}
\]

\[
\rho(t_n) = \rho(t_{n-1}) + \Delta \rho(t_n)
\]

The Raman-Kerr integral \(\int_{-\infty}^{t} R(t - t')|\varepsilon(t')|^2\ dt'\) is recalculated at the beginning of each step in a similar manner.

Figure 11. Flowchart of filament simulation program based on solving the NLSE by the split-step method.

The process described in the paragraphs above and in Figure 11 is coded in Matlab. The Hankel transform algorithm adopted has been developed and written by Guizar-Sicairos.
and Gutiérrez-Vega [118]. Data is not saved at every step, but at a pre-determined propagation interval. Key data saved are the complex 2D matrix $\varepsilon(r, t)$ and the real 2D matrix $\rho(r, t)$. Secondary data – maximum on-axis intensity, energy contained in the profile and the fluence half-maximum position – are also saved for easy retrieval.

2.2.2 - Comparison of Simulation with Measured Profiles

To verify that the simulations produce results that are consistent with experimental measurements, the NLSE solver code was used to simulate filaments corresponding to the conditions of the short and medium distance measurements described in Chapter 2.1.2. The only difference was that the pulse energy was limited to 1 mJ, as simulations of single filaments reflect experimental measurements better when the simulated peak power does not exceed $P_{cr}$ by too large a margin. The long distance measurement was not simulated as the longer duration of the chirped pulse requires significantly more computational resources.
2.2.2.1 - Short Distance Comparison

Figure 12. Comparison of short distance filament measurements with simulation.

Figure 13. Detailed simulation results of short distance filament. The top image is a map of the fluence of the filament, the middle plot shows the peak intensity and plasma density along $z$, and the bottom image maps the on-axis intensity of the filament in time as it propagates.
Figure 12 and Figure 13 show that there is good agreement in the position and length of the filament – between $z = 2$ m and 2.2 m. However, the simulations predict that the fluence profile takes on a donut shape in the middle of the filament (Figure 13, top), which accounts for the larger FWHM beam size. In the measured profiles, the fluence of the rings remained at a fraction of that of the central peak. A point to note is how the effect of pulse splitting begins to appear at the end of the filament, but the linear divergence of the beam terminates the filament before the split pulses can propagate (Figure 13, bottom).

2.2.2.2 - Medium Distance Comparison

Figure 14. Comparison of medium distance filament measurements with simulation.
The size of the filament predicted by simulation agrees well with the measurements, though the measurements show a gentler evolution of the beam size compared to the simulation. The ring structure that was observed in the short distance case is absent here in both the measurements and simulation. What is not revealed in the measurements but clearly seen in the simulation is the pulse splitting effect just before 11 m, shortly after the collapse of the beam and the refocusing event.

2.2.2.3 - Discussion

The output of the simulation shows all the characteristics that are known to be typical for laser filaments. For example, the clamped intensity and the plasma density in the filament fall within the expected range of values, and the pulse splitting and refocusing effects have been reproduced. The comparisons with the measured fluence profiles in real filaments
allow for an extra level of verification of the simulated results. Some discrepancies exist between the simulated and measured profiles, possibly because only the essential physical effects have been included in the current NLSE model. Effects such as higher-order dispersion, space-time focusing and self-steepening have been excluded for simplicity. It has been shown that the fine accuracy of the simulation can be affected by the omission of these and other terms [119–121].

2.3 - Summary

The grazing incidence imaging system has been shown to produce reliable measurements of the filament fluence profile, and its portability allows for measurements at arbitrary positions along the filamenting beam. The simulation of a filamenting laser pulse through the resolution of the NLSE by the split-step method has also produced results that are consistent with other published data, both simulated and experimental. These tools will contribute to the laboratory’s effort to unravel the complex physics behind laser filaments and their interaction with materials, as well as to develop cutting-edge filament-based applications.
CHAPTER 3 - DIFFERENT REGIMES OF FILAMENTATION – EXPERIMENTS IN SOLIDS

Filamentation can occur in a very broad range of conditions; if the medium is effectively transparent to the laser pulse, and the pulse has sufficient energy to induce self-focusing in the medium, filamentation could potentially take place. However, depending on the properties of the medium and the laser pulse, the filament that forms can behave very differently. Many parameters potentially affect the filamentation process, and it is important to understand the contribution of these parameters so as to better control the process. For example, imposing an initial chirp in the pulse has been shown to be useful for influencing the position and length of the filament, as well as the supercontinuum generated [111, 122–124].

As part of this dissertation, we have examined the effects of anomalous dispersion and the influence of different multi-photon ionization regimes on laser filaments. However, these studies could not be effectively performed with air as the propagation medium, as the use of Ti:sapphire laser and optical parametric amplifier (OPA) sources limited the range of wavelengths and/or peak powers that was available.

- In air, the anomalous dispersion region resides above 3 µm, while the longest wavelength achievable with the OPA is around 2.5 µm. Moreover, Shim et al. have performed a theoretical study to show how the 3.1 µm anomalous dispersion window in air is too narrow and not suitable for the study of filamentation [125], so high peak power sources further in the mid-IR are needed.
• The ionization potential (based on $O_2$) is 12 eV. To be able to explore multi-photon ionization effects from the 2- to 5-photon ionization regime, pulses with central wavelengths between 206 nm to 516 nm would be necessary.

The experiments described in this chapter were therefore carried out in condensed media. Fused silica, which exhibits anomalous dispersion at wavelengths above 1.26 $\mu$m [126], was used as the propagation medium for the first experiment, while zinc selenide (ZnSe), which has a bandgap of 2.67 eV [127], was used in the second.

3.1 - Supercontinuum Generation in the Anomalous Dispersion Regime

The study of laser filamentation has commonly been carried out in the normal dispersion regime (for example, at 800 nm in air). The Kerr-induced spectral broadening in the pulse introduces new frequencies of higher (lower) energy in the trailing (leading) edge of the pulse. When propagating in a medium with normal dispersion, the pulse will tend to split as dispersion effects pull the new frequencies in opposite directions. This pulse splitting effect is responsible for rapid dissipation of energy after the formation of the filament, leading to more pronounced defocusing and refocusing events.

Figure 16. Diagram illustrating the effect of dispersion on a Kerr effect spectrally broadened pulse.
Analogous to a temporal soliton in an optical fiber, anomalous dispersion can help to sweep the new frequencies generated in a filament back towards the center of the pulse, and keep the pulse propagating as a “light bullet” for a much longer duration and distance. Due to the broadened spectrum, the pulse can even be self-compressed to a duration shorter than its initial duration. Using a planar glass waveguide, Eisenberg et al. showed the first demonstration of simultaneous spatial and temporal focusing by Kerr effect in the anomalous dispersion regime [128]. Moll and Gaeta later demonstrated in BK7 that filaments in the anomalous dispersion regime extend further than in the normal dispersion regime [129]. Pulse self-compression by filamentation in the anomalous dispersion regime of fused silica was recently shown by Durand et al. [63].

Figure 17. Images (left) and spectra (right) of filaments in fused silica. (a) shows a filament in the normal dispersion regime, while (b) and (c) are in the anomalous dispersion regime. White arrows indicate refocusing events. The filaments in the anomalous dispersion regime show greater uniformity and persist for longer. The spectra show the expected broadening around the central wavelength for normal dispersion (red) and the blueshifted spectral peak for anomalous dispersion (black). Source: [63]

Saliminia et al. [130] and Faccio et al. [131] have measured the supercontinuum generated by filaments propagating in anomalously dispersive media, and have noted the appearance of a distinct blueshifted spectral peak that is separate from the broadened spectrum.
around the pulse central wavelength. The description of the far-field spectra of filaments as stationary solutions of the NLSE in the form of X- and O-waves (in the normal and anomalous dispersion regimes respectively) has been previously established [132, 133]. Faccio et al. accurately described the blueshifted peak as the fishtail-like feature that accompanies an O-wave [131]. Smetanina et al. made similar observations in fused silica and explained the emergence of the separate blueshifted peak as a combination of the strong spectral broadening towards the high frequencies and destructive interference effects [134–136]. Based on our experimental and numerical data, we offer an alternative explanation of the blueshifted peak that complements those mentioned above, using the effective three-wave mixing (ETWM) model developed by Kolesik et al. [137, 138].

3.1.1 - Experimental Setup and Results

The source used in our experiment was an OPA (Opera F, Coherent) pumped by 1.1 mJ, 100 fs pulses at 800 nm (Spitfire, Spectra-Physics). The central wavelength of the filamenting pulses was varied from 1.2 to 2.4 μm, and the beam was focused with a
A 41 mm lens inside a 4 cm long fused silica rod (Hexagonal light pipe, Edmund). The on-axis spectrum at the output of the rod was picked up by an optical fiber and measured by a spectrometer. To cover the full spectral range of interest, three spectrometers were used [Ocean Optics HR 4000 (298-756 nm), HR 4000 (711-889 nm) and NIRQUEST (853-2571 nm)] and calibrated using a NIST-calibrated tungsten lamp. The energy of the pulses was controlled by a variable reflectivity wheel. For each wavelength, the pulse energy was kept to the minimum required to observe a stable and radially symmetric conical emission pattern. This was measured to be several μJ for the shorter wavelengths, and between 10-20 μJ for 2.3 μm and 2.4 μm.
Figure 19. Supercontinuum spectra with different initial wavelength $\lambda_0$ indicated on the left (Source: [139]). Dashed lines in the plots help to guide the eye along the peaks of $\lambda_0$ (red) and the blueshifted peaks (blue). Parts of the spectra below 1 $\mu$m has been magnified by the factor indicated. On the left are images of the supercontinuum taken with a commercial digital SLR for different $\lambda_0$.

The measurements and images shown in Figure 19 show a clear decrease in the blueshifted peak’s wavelength as the initial wavelength $\lambda_0$ increases. The images captured by a digital SLR camera, which is not sensitive to wavelengths above 1 $\mu$m, also exhibit a clear shift of colors from yellow to blue with increasing $\lambda_0$. This is the first time that such a trend has been observed for filaments in the anomalous dispersion regime.

3.1.2 - Effective Three-wave Mixing Model

To interpret this trend, we have to first understand the features of the far-field spectrum of the filament in terms of the ETWM model [137, 138]. In summary, the ETWM model treats
the collective complex refractive index perturbation due to nonlinear effects in the filament as a traveling material polarization wave. The polarization wave has a frequency of \( \Omega = \omega - \omega_0 \), where \( \omega_0 \) is the frequency of the initial pulse. When the polarization wave interacts with the filamenting pulse, a scattered wave with frequency \( \omega \) and transverse wave vector component \( k_\perp \) is generated. The associated phase-matching condition for the scattered wave is

\[
\Delta k = k_0 + \frac{\Omega}{v_p} - k_x(\omega, k_\perp) \approx 0 \tag{17}
\]

where \( v_p \) is the velocity of the peak of the pulse (which can be different from the group velocity \( v_g \)) and \( k_x \) is the longitudinal wave vector component of the scattered wave. The scattered wave also has to fulfill the dispersion relation of the medium,

\[
k^2_\perp + k^2_z = k^2(\omega) \tag{18}
\]

By combining Equations (17) and (18), the loci of the supported new frequencies are therefore defined by

\[
k_\perp = \pm \sqrt{k^2(\omega) - (k_0 - \Delta k + \alpha \Omega)^2} \tag{19}
\]

where \( \alpha = 1/v_p \).
3.1.3 - Comparison of Measurements with ETWM Model

Figure 20. (a) shows the simulated temporal profile of a $\lambda_0 = 1.9 \, \mu m$, 70 fs, 7.2 $\mu J$ pulse propagating in fused silica. The black line indicates the slope for calculating $v_p$. (b) shows the simulated far-field spectrum of the filament, with the dashed white lines indicating the loci of the frequencies generated by ETWM as predicted by Equation (19). (c) shows the measured far-field spectrum of a $\lambda_0 = 1.9 \, \mu m$, 40 fs, 26 $\mu J$ pulse after propagating in 3 cm of fused silica, with the ETWM loci indicated by the white lines. Source: [139]

When simulated data of the filament is available, $\alpha$ can be deduced from the evolution of the temporal profile of the pulse as it propagates. In Figure 20(a), the simulated profile of pulse shows that it has a subluminal velocity beyond $z = 2 \, cm$. Since the reference frame moves with the pulse at its group velocity $v_g$, $\alpha$ can be deduced by $\alpha = \frac{1}{v_g} + \frac{dt}{dz}$. Figure 20(b) shows the ETWM loci with $\alpha = 1.0002/v_g$ as determined from Figure 20(a). In experimental results, $\alpha$ can be used as a fitting parameter; Figure 20(c) shows a fit of the ETWM loci with the measured far-field spectrum using $\alpha = 1.0035/v_g$. The position of the fishtail-like portion of the far-field spectrum, which accounts for the blueshifted peak, can therefore be accurately predicted by ETWM.
Equation (19) was solved for different $k_0$ and $v_g$, with $\alpha = 1.0002/v_g$, corresponding to the range of $\lambda_0$ used. The predicted positions of the blueshifted peak agreed well with the measured positions for wavelengths 1.5 $\mu$m and longer. The deviation from theory for wavelengths below 1.5 $\mu$m can be explained by the proximity to the zero-dispersion wavelength, and therefore the transition between features that are characteristic of filaments in the normal dispersion regime versus in the anomalous dispersion regime. The ETWM theory predicts X- and O-wave structures equally well, but the difference in the temporal evolution of the pulse in the two dispersion regimes means that $\alpha$ can no longer be approximated by a single value. Once pulse splitting, a typical behavior in the normal dispersion regime, takes place, a separate value of $\alpha$ has to determined for each split pulse, consequently producing two sets of loci that form the X-wave structure.
3.1.4 - Summary

This study has allowed us to explain the appearance of the separate blueshifted spectral peak for a filament in the anomalous dispersion regime as the manifestation of phase-matching effects in the supercontinuum generation process. The ETWM model provides a quick and accurate way to predict the position of the blueshifted peak, and the predictions fit well with both simulated and experimental data.

3.2 - Formation of Filaments in Different Multi-photon Absorption Regimes

Solid state near-IR laser systems, most prominently Ti:sapphire-based systems, are currently the most accessible type of source for generating ultrafast pulses with the high peak powers necessary for creating filaments in air and other common gases. As the photon energy of these sources is in the range of ~1.2-1.6 eV and the ionization potential of the common gases (N\textsubscript{2}, O\textsubscript{2}, CO\textsubscript{2} and Ar) resides in the range ~12-16 eV, filamentation studies have been largely limited to a similar MPI regime. With the development of novel high power ultrafast sources in the mid-IR and the UV, filamentation studies in very different ionization regimes will become more widespread \cite{29, 140–143}.

There is significant interest in generating filaments with pulses of shorter wavelengths, as such filaments have the potential to confine more energy in their cores \cite{140} and to lose less energy to conical emission during propagation \cite{29}. However, a thorough study of filamentation in the low-order MPI regime has yet to be conducted. For condensed media, there have been studies to investigate the influence of the material band gap, \( E_g \), on
supercontinuum generation. Brodeur and Chin have shown that materials with larger band gaps generate broader supercontinua when pumped with the same laser wavelength [144, 145]. Nagura et al. carried out a similar and more elaborate study, and deduced that a threshold of $E_g/h\nu_0 = 2$ exists for supercontinuum generation due to the competition between SPM and 2-photon absorption [146]. As filamentation and supercontinuum generation are intricately linked, this implies that the interplay between Kerr-related effects and multi-photon processes can impact the formation of filaments. Couairon and Mysyrowicz have suggested that for filamentation in gases, if $U_i < 3h\nu_0$, gradual attenuation of the pulse by 2- and 3-photon absorption would inhibit the formation of a well-confined filament [6]. In previously published data, the focus was on the supercontinua and there was minimal attention paid to the filaments' properties. The filamentation threshold suggested by Couairon and Mysyrowicz has therefore not been directly observed.

In this study, we explore this threshold by observing the evolution of filament structures generated by near-IR pulses between 800 nm ($h\nu_0 = 1.55$ eV) to 2.4 $\mu$m ($h\nu_0 = 0.52$ eV) propagating in a zinc selenide (ZnSe) crystal. ZnSe has a relatively small band gap of $E_g = 2.67$ eV [127], and the wavelengths available allow the range of multi-photon absorption (MPA) from 2-photon to 6-photon to be investigated. ZnSe is also normally dispersive and exhibits minimal change in $n_0$ and $n_2$ within the entire spectral window of interest [147, 148], making it an ideal medium for isolating the impact of MPA on filament formation.
3.2.1 - Experimental Setup

Figure 22. Diagram of experimental setup. Source: [149]

The source used in the experiment was an OPA (Palitra, Quantronix) pumped by 0.5 mJ, 50 fs, 800 nm pulses delivered by a kHz CPA laser system. For all the wavelengths used, the pulse energy (6-20 μJ) was sufficiently high to ensure that the peak power was largely superior to $P_{cr}$ (between 60 kW and 400 kW [148]) in ZnSe, evident in the multi-filament bundles generated (Figure 24, right). The pulses were focused on the front surface of the ZnSe crystal by a $f = 200$ mm lens. Images of the filaments were recorded from the side by a digital camera coupled with a microscope objective. The supercontinuum generated was also recorded after diffusion off a white screen by two spectrometers (Ocean Optics HR 4000 and NIRQUEST) to cover the spectral range of 0.3-2.5 μm.
3.2.2 - Observations and Discussion

Figure 23. Spectra of the laser before (black) and after (red) the ZnSe crystal. The spectra shown are for $\lambda_0 = 800$ nm (solid lines) and $\lambda_0 = 1200$ nm (dashed lines). Inset: the bright blue spot on the input surface of the crystal for $\lambda_0 = 800$ nm. Source: [149]

The 2-photon absorption regime was first examined using $\lambda_0 = 800$ nm. At this wavelength, there was no visible formation of filaments in the ZnSe crystal. At the same time, the spectrum at the output of the crystal exhibited no significant broadening. On the input surface of the crystal, a bright blue spot due to 2-photon luminescence could be observed. This is in agreement with other published results showing that supercontinuum generation and filamentation do not occur in the 2-photon absorption regime. The same situation was observed when $\lambda_0$ was switched to 600 nm.
Figure 24. Left: Spectra of photoluminescence measured at different laser wavelengths. Right: Images of filaments taken from the side of the crystal for different laser wavelength and MPI regime. Source: [149]

When pulses of longer wavelengths are used, bright blue strings of light were seen to appear in the ZnSe crystal. For filamentation in gases, plasma emission from the relaxation of excited gas ions, molecules and atoms is frequently used as a proxy for identifying and characterizing the filaments. In a solid medium, the photoluminescence can serve the same purpose. To verify that the blue light strings are indeed from photoluminescence effects, the digital camera in the setup was replace by a 0.5 m Czerny-Turner spectrometer (2500i, Princeton Instruments) with a 600 lp/mm grating, coupled to an iCCD camera (iStar 720, Andor). The spectrum of the light strings was seen to be invariant with the source wavelength (Figure 24, left), and corresponded well with the band edge emission wavelength of ZnSe. We concluded therefore that the blue light strings were a reliable representation of filaments in the crystal.
The images in Figure 24 show the evolution of the filament structure as the laser wavelength was increased, moving from the 3-photon absorption regime to the 6-photon absorption regime. In the 3-photon absorption regime ($\lambda_0 = 1.15 \mu m$ and $1.2 \mu m$), the photoluminescence appears in two distinct zones. At the front surface of the crystal where the geometrical focus of the beam is, a bright conical zone is observed. This is unlike the 2-photon absorption case where the emission is limited to the surface of the crystal. The photoluminescence then stops before reappearing about 0.5 cm into the crystal and persists for about 1 cm, in the form resembling a multi-filament bundle. In the 3-photon regime, the absorption is still significant near the geometrical focus where the intensity is high, and the MPA effect is sufficient to arrest the formation of filaments. Only after the beam diverges and the intensity, therefore MPA, has reached a low enough level can the dynamic balance between Kerr self-focusing and the relevant arresting effects (in this case possibly MPA and dispersion in addition to defocusing by free-electrons) be established. This demonstrates that it is possible to form filaments in the 3-photon absorption regime, though their formation may be easily disrupted by external focusing conditions.

Figure 24(c) and (d) illustrate the transition from the 3- to 4-photon absorption regime. At $\lambda_0 = 1.4 \mu m$, the spectrum of the pulse is fairly evenly divided between the 3- and 4-photon absorption region. MPA is therefore still strong enough around the geometrical focus to arrest the formation of filaments, and the initial blue photoluminescence cone is observed. However, the subsequent filament bundle forms much closer to the initial collapse of the
beam at the geometrical focus, and contains more and longer filaments than in Figure 24(a) and (b). At $\lambda_0 = 1.7 \mu m$, the spectrum resides well into the 4-photon absorption region, and MPA is sufficiently weak to allow a continuous bundle of filaments to form upon the entry of the pulse at the front surface of the crystal. As the wavelength was switched into the 5- and 6-photon absorption region, the photoluminescence at the front surface decreases in intensity, indicating weaker MPA. A continuous filament bundle still forms in the bulk of the crystal. In the 6-photon absorption regime, the bundle appears to shrink in diameter but grow in length and uniformity. This may be an indication of the formation of fewer but more stable filaments and a lower tendency for the beam to be affected by modulational instabilities.

Figure 25. Measured amount of spectral broadening at the different laser wavelengths as a function of the number of photons required to overcome $E_g$. Source: [149]

In addition to the direct visual observation of the filaments, the spectral broadening effect at the output of the crystal was also examined. The spectra of the pulse for the different wavelengths were taken with and without the crystal (propagation in air), and the amount of broadening $\Delta \omega$ for each wavelength was calculated based on the different spectral
widths (see Figure 23 for examples at 800 nm and 1.2 μm). The trend is plotted as a function of the number of photons required to overcome $E_g$, and it is consistent with the observations made by Brodeur and Chin at a fixed wavelength filamenting in different materials. $\lambda_0 = 1.4$ μm exhibits an intermediate amount of spectral broadening due to the transition between 3- and 4-photon MPA. The spectral broadening measurements are also in good agreement with the visual filament observations – supercontinuum generation and filamentation do not occur at the 2-photon absorption level, and grow in prominence as the MPA level increases.

3.2.3 - Summary

In conclusion, we have isolated the impact of the MPA regime on the formation of filaments in ZnSe. This is the first time direct visual observation of the filament behavior has been made while switching between different MPA regimes, and the observations confirm that filaments can only form when MPA involves at least three photons. In the 3-photon absorption regime, even though filamentation is possible, MPA retains a strong arresting effect and may limit the extent to which filamentation can take place, leading to fewer and shorter filaments observed. As the number of photons required for MPA increases, absorption becomes less able to arrest filamentation and more prominent and stable filament structures can form.
CHAPTER 4 - DIFFERENT REGIMES OF FILAMENTATION – EXPERIMENTS IN GASES

While experiments with filaments in solid media can provide new insights into the science of filamentation, the findings may not immediately benefit many applications of laser filaments, which frequently involve propagation through air. For the potential users of filamentation, it is often important that the source is a readily available and mature laser system. Therefore the study of filamentation in air and gases with pulses in the NIR will likely be the most impactful and provides the greatest benefit towards the deployment of filamentation in the field.

Even though many aspects of filamentation science has been carefully investigated over the years, the complexity of the nonlinear effects involved is such that we still do not have a complete picture of how all the physical processes work together. In this chapter, we describe three investigative studies conducted on filaments in air and other gases to obtain a better understanding of how different initial conditions can impact the outcome of filamentation. The first examined how geometrical focusing can effectively bring about two different regimes of filamentation. The second looked at how the initial polarization of the pulse can affect the filaments formed. The third attempted to identify some of the features of filaments generated by UV pulses and how they differ from those generated by NIR pulses.
4.1 - The Effects of Geometrical Focusing: Linear-focusing and Nonlinear-focusing Regimes of Filamentation in Gases

Depending on the nature of the investigation conducted, filaments have been formed in a variety of initial conditions. One key parameter that inevitably differs from one experiment to another is the initial geometrical focusing condition. Out in the field, large laser systems are frequently used as the laser source, and the laser output is often a collimated beam or weakly focused with large telescopes [111, 124, 150]. Conversely, in the laboratory, filaments are typically initiated from smaller beams and using focusing optics with focal lengths of a few meters or less [5, 102, 103, 151]. Two significant consequences of tight geometrical focusing have been reported: the plasma density in the filament is increased [54, 152–155], and the supercontinuum generated (for a 800 nm pulse) extends into the visible but not towards the IR [62, 156, 157]. The dominant effect of geometrical focusing over Kerr self-focusing and plasma defocusing has generally been cited as the explanation for these differences. However, the transition from a loosely focused, “typical” filament to a tightly focused filament has not been examined, and the role of geometrical focusing on the filamentation process has not been systematically studied.

Due to the complex nature of the physical processes involved in filamentation, it is usually difficult to predict the behavior of a filamenting beam without resorting to lengthy numerical simulations. One notable exception is Marburger’s formula (Eqs. (3) and (4)), which provides a quick approximation of where a filament begins to form given its initial parameters. We propose a new analytical tool that helps to differentiate between filaments formed in two distinct regimes based on geometrical focusing conditions.
4.1.1 - Analytical Method to Determine the Transition Between Regimes

To perform an analytical study of filament behavior, only the key physical processes – diffraction, Kerr self-focusing (KSF) and plasma defocusing – are considered. In most gases, other effects such as MPA, plasma absorption and dispersion play secondary roles that have limited impact on the defining characteristics of the filaments. Braun et al. [5] carried out a similar analysis on the three key physical processes that predicted fairly accurately the clamped intensity and plasma density in a filament. We adopt a similar approach, replacing the diffraction term by another that describes a focusing Gaussian beam.

Figure 26. Illustration of a focusing Gaussian beam and the parameters for calculating the wavefront sag, \( s \).

In this analysis, the wavefront sag of the beam is used as the indicator of the effects of KSF and plasma defocusing on a focusing Gaussian beam (Figure 26). The default wavefront sag at any position \( z \) of a focusing Gaussian beam (with focal distance \( f \) and \( z = f \) being the focal position), \( s_G \), is given by the formulae
The Rayleigh distance is given by

\[ w(z) = w_0 \sqrt{1 + \left(\frac{z - f}{z_R}\right)^2} \]  \hspace{1cm} (20)

\[ R(z) = (z - f) \left(1 + \left(\frac{z_R}{z - f}\right)^2\right) \]  \hspace{1cm} (21)

\[ s_G = R(z) - \sqrt{R(z)^2 - w(z)^2} \]

\[ \approx \frac{w(z)^2}{2R(z)} \]  \hspace{1cm} (22)

\[ \approx \frac{w_0^2}{2z_R^2} (z - f) \]

The Rayleigh distance is given by \( z_R = \frac{\pi w_0^2}{\lambda_0} \) and the beam waist is approximated as \( w_0 \approx \frac{\lambda_0 f}{\pi w(0)} = \frac{\lambda_0}{\pi \text{NA}} \). This approximation is valid when \( \text{NA} \cdot w(0) \gg \frac{\lambda_0}{\pi} \), which is the case for 800 nm pulses that are 1 mm or larger in diameter. The first approximation in Equation (22) requires that \( \lambda_0/\pi w_0 \ll 1 \), i.e. \( \text{NA} \ll 1 \), which is true for the focusing conditions of interest in this study. The sag contribution of KSF and plasma defocusing is approximated by the cumulative optical path difference between the center and the edge of the beam, such that

\[ s = -\int_0^z \Delta n(z') \, dz' \]  \hspace{1cm} (23)

For KSF, this is similar to the calculation of the B-integral, with \( \Delta n(z) = n_2 I_0(z) \). For a geometrically focusing beam with numerical aperture (NA) typical of laboratory conditions (e.g. \( \text{NA} \simeq 10^{-3} \)), the beam propagates very similarly to a Gaussian beam for most of the distance prior to filamentation. The Gaussian beam approximation \( I_0(z) \approx \frac{2P_0}{\pi w(z)^2} \) can
therefore be made, where \( P_0 \) is the peak power of the pulse. For a Gaussian pulse, the peak power is calculated by \( P_0 = \sqrt{\frac{41n2E}{\pi \tau}} \) where \( E \) is the pulse energy and \( \tau \) is the FWHM pulse duration. Solving Equation (23) for KSF to obtain its sag contribution gives

\[
s_K = \frac{2n_2 P_0 z_R}{\pi w_0^2} \left( \tan^{-1} \frac{z - f}{z_R} + \tan^{-1} \frac{f}{z_R} \right)
\]

For plasma defocusing, the index change is given by \( \Delta n(z) = \frac{\rho(z)}{2\rho_c} \). By assuming that the MPI rate can be used to quantify plasma generation in a filament, the plasma density can then be approximated by \( \rho(z) = \sigma_K \rho_{nt} \tau(I_0(z))^K \). The other ionization parameters are as described in Chapter 1.1.2. The sag contribution from plasma defocusing is therefore deduced to be

\[
s_p = \frac{\sigma_K \rho_{nt} \tau z_R}{2 \rho_c} \left( \frac{2P_0}{\pi w_0^2} \right)^K \left( \frac{2K - 2)!}{(2^{K-1}(K-1)!)} \left( \frac{h_K(\frac{z-f}{z_R}) + h_K(\frac{f}{z_R})}{z_R} \right) \right)
\]

where

\[
h_K(x) = \tan^{-1} x + \sum_{n=1}^{K-1} \frac{(2^n n!)^2}{2n(1+x^2)^n} \frac{x}{2n(1+x^2)^n}
\]

Just as explained in Chapter 1.1.2, the plasma in the filament is considered to originate from just the oxygen in air [18]. Additional calculations to include the contribution of nitrogen to the value of \( |s_p| \) showed that it has negligible effects on the outcome of this analysis.
The wavefront sag contributions of geometrical focusing, KSF and plasma defocusing can be plotted together to track their relationship with propagation distance (Figure 27). As nonlinear effects are weak when the intensity is low, both $|s_K|$ and $|s_p|$ are negligible for most of the distance before the focal point. Near the focus where the intensity is high, $|s_K|$ and $|s_p|$ increase rapidly. It is clear that the effect of KSF builds up more gradually, while plasma defocusing sets in more abruptly (once $I_0 > 10^{13}$ Wcm$^{-2}$). The positions $z_K$ and $z_p$ are defined as the positions where $|s_K| = |s_G|$ and $|s_p| = |s_G|$ respectively. These positions are considered to be where the corresponding nonlinear effect has an effect on the wavefront that is non-negligible compared to geometrical focusing. In high-NA conditions (Figure 27a), the effect of KSF has limited distance to build up, therefore $z_p < z_K$. The opposite occurs for high-NA conditions (Figure 27c). There is therefore a transition focusing condition where $z_p = z_K$, where the impact of KSF and plasma defocusing gain
significance at the same time (Figure 27b). For the initial beam parameters used in Figure 27, this transition occurs at $\text{NA} = \frac{w(0)}{f} = 4.25 \times 10^{-3}$.

4.1.2 - Numerical and Experimental Verification of the Analysis

Figure 28. Numerically determined half-max beam sizes based on the fluence profiles. For each initial focal distance, simulations were carried out using the full NLSE (green solid line), NLSE without plasma effects (blue dashed lines) and NLSE without Kerr effect (red dash-dot lines). The theoretical Gaussian beam profile (black dotted line), as well as the calculated positions of Marburger’s self-focusing distance, $z_K$ and $z_p$, are also indicated for comparison.

The first step to determining the soundness of the proposed transition between high-NA and low-NA filaments is to validate the definition of $z_K$ and $z_p$. Since these positions were defined based on a linearly propagating Gaussian beam, their significance should be evident when comparing a Gaussian beam that is undergoing transformation due to just the relevant effect (KSF or plasma) with linear Gaussian propagation. Three sets of simulations were carried out – one with the full NLSE as described in Chapter 2.2.1, and one each with
all plasma related terms and Kerr effect related terms removed. The initial conditions used were identical to those for the calculations in Figure 27, and the results are shown in Figure 28.

The simulations without plasma effects show that a self-focusing beam will collapse to a singularity as the position as predicted by Marburger's formula. $z_K$ is always located before this theoretical collapse position; this makes sense as $z_K$ indicates the position where KSF becomes significant compared to geometrical focusing, and this has to happen before the collapse of the beam can occur. The beam from the simulations without Kerr effect follows the Gaussian profile exactly up to a point where it abruptly defocuses, indicating that the rapid build up of plasma from MPI has occurred. The position where defocusing occurs corresponds very well with $z_p$, thereby validating its definition.

Furthermore, for high-NA conditions ($f < 1$ m, Figure 27a and Figure 27b), the simulation results without Kerr effect show a high level of similarity with the full NLSE results. This similarity is less striking at the transition ($f = 1$ m, Figure 27c) and is absent in the low-NA case ($f > 1$ m, Figure 27d). This suggests that KSF plays a noticeable but minimal role compared to geometrical focusing and plasma defocusing in the high-NA conditions, while it is critical for deducing the correct behavior of a filamentating beam in low-NA conditions. The transition where $z_K = z_p$ therefore defines a real change in the balance of physical processes that are important for filamentation. To identify the two regimes by the focusing
mechanism that dominates in the filamentation process, we label the high-NA condition the linear-focusing regime and the low-NA condition the nonlinear-focusing regime.

4.1.3 - Differences between Linear-focusing and Nonlinear-focusing Regimes

The beam profiles from simulation shown in Figure 28 are but one indication that a change in physical regime has taken place once the condition $z_K = z_p$ has been crossed. Given that the contribution of Kerr effect is different in the two regimes, other differences in the characteristics of the filaments formed in the two regimes are expected. Verifying such differences also strengthen the argument that the transition and the two regimes have been accurately determined.

4.1.3.1 - Filamentation Start Position

Figure 29. The measured and simulated positions of where filamentation begins, plotted together with the Marburger collapse position and $z_p$ for comparison. The measurements were carried out for two different pulse energies, (a) 3 mJ and (b) 5 mJ, while the corresponding simulations and calculations were carried out with (a) 0.8 mJ and (b) 1.33 mJ.

Marburger’s formula and the formula for the modified self-focusing distance for a geometrically focusing beam (Eqs. (3) and (4)) have often been used to estimate the
position where a filament begins to form. The observations in Figure 28 suggest that $z_p$ may be a better approximation for the starting position of the filament in the linear-focusing regime. To test this hypothesis, the starting positions of filaments formed by different focusing lens were measured in the laboratory. This was done by noting the positions where plasma emission from the filaments becomes visible to the naked eye in a darkened laboratory. The starting positions of the simulated filaments were taken to be where the plasma density first reaches a plateau (see Figure 30). In Figure 29, it is clear that the collapse position predicted by Marburger is further than the starting position of the filament. This is expected since plasma would be generated and filamentation occur before the beam can completely collapse. Nonetheless, for shorter focal distances, the difference between the actual filamentation start position and Marburger’s collapse position becomes greater (relative to the geometrical focal distance), and both measured and simulated filament start positions become better approximated by $z_p$. The simulations and analytical calculations were carried out at pulse energies that were $\sim 3x$ lower than energies used in the experimental measurements. This was justified by the fact that the critical power for self-focusing has been shown to increase for pulses shorter than 200 fs [158], and filamentation was effectively not observed experimentally for pulses with peak power $P_0 \leq 3P_{cr}$. 
4.1.3.2 - Trends in Filament Characteristics

Figure 30. Plots of simulated peak intensity and plasma density in the filaments for different initial focal distances (indicated in meters in the legend). Plots in the linear-focusing regime and transition point are in solid lines, while plots in the nonlinear-focusing regime are in dashed lines. Initial beam parameters are identical to Figure 27.

The trends in the simulation results for the peak intensity and plasma density in the filaments are noticeably different. In the nonlinear-focusing regime, the peak intensities plateaued at very similar values, which is consistent with the concept of a clamping intensity in filaments. The plasma densities stayed at values just over $10^{16}$ cm$^{-3}$, which is in the expected range for filaments at 800 nm in air. However, after transitioning into the linear-focusing regime, the peak intensities increased beyond the clamped value of the nonlinear-focusing cases, and there was a rapid increase in plasma densities with higher initial NA. These are characteristics reported by Prem Kiran et al. [159] and Théberge et al. [54] for tight focusing geometries, and here we observe the appearance of these changes as we transition from nonlinear-focusing to linear-focusing regime. In the linear-focusing regime, the peak intensity prior to filamentation was also observed to increase at similar
rates over the normalized distance, which is indicative of geometrical focusing with minimal influence by KSF.

4.1.3.3 - Spectral Broadening
Differences in the supercontinuum generated have been reported to be the most distinctive change observable between the two regimes. Published results show that tight focusing geometries produce spectral broadening primarily towards the visible wavelengths [62, 156, 157], while the “typical” filament is known to produce a supercontinuum that stretches both into the visible and well into the infrared wavelengths [20, 160]. This is also an indication of the secondary role Kerr effect plays in the linear-focusing, or high-NA, regime, as SPM due to the Kerr effect will broaden the spectrum in both directions, while spectral broadening due to plasma effects is limited to towards higher frequencies [9, 62].

The supercontinuum spectrum after filamentation was measured in the laboratory by placing a white diffusing screen after filamentation has terminated, and collecting the diffused light into a fiber-coupled spectrometer (Ocean Optics HR2000 for visible wavelengths and USB2000 for NIR wavelengths). The spot on the screen was kept small so that all the light could be coupled into the large core multi-mode fiber, but large enough to ensure that no damage was observed on the screen after prolonged exposure. To ensure that all the light was captured, the measured spectra were observed to stay the same even when the fiber tip and collection optics were slightly tilted and shifted in either direction.
For the numerical results, the spectra of the pulses at the end of the simulation were extracted and radially integrated to obtain the spectra in the whole beam.

Figure 31. Spectra after filamentation from simulations (a,b) and measurements (c,d). The initial beam conditions were as described in Figure 27 for both simulation and experimental data, while pulse energies were 0.8 mJ for the simulations and 3 mJ in the experiments. The initial geometrical focal distances are indicated in the legends. The transition at \( f = 1 \) m are plotted in all four cases for easy comparison. The original laser output spectrum is plotted as the black dashed lines.

Both numerical and experimental results exhibited the expected trend – little to no spectral broadening in the IR in the linear-focusing regime. In the linear-focusing regime, the broadening in the visible also decreased as the focal distance increased closer to the transition point, as plasma densities decreased. Once into the nonlinear-focusing regime, as the focal distance increased, the broadening could be observed to increase in both directions, as SPM took over and became more significant the further from the transition point.
4.1.3.4 - Temporal Pulse Splitting

Figure 32. Evolution of the on-axis temporal profile of the pulses, obtained from the full NLSE simulations and the simulations without Kerr effect.

In the linear-focusing regime, the similarities between the simulation results with the full NLSE and the NLSE without Kerr effect could be observed not just in the beam size (Figure 28a,b), but in the temporal evolution of the pulse as well (Figure 32a,b,e,f). Once filamentation began, the peak of the pulse appeared to travel faster than the group velocity, as plasma defocusing and absorption affected primarily the rear of the pulse. After filamentation ended and plasma density decreased, some energy in the rear of the pulse returned to the propagation axis. With KSF, more energy was refocused on-axis after the filament (Figure 32a,b), but the impact on the overall temporal profile of the pulse was minimal.

In the nonlinear-focusing case (Figure 32d), the full NLSE simulation showed pulse splitting taking place, with the split pulse persisting for a significant distance. Pulse splitting in
filamentation is understood to be due to a combination of SPM, dispersion and plasma defocusing [102, 161, 162]. It can therefore not occur in the linear-focusing regime due to the limited impact of the Kerr effect. For filamentation at 400 nm in water, Jarnac et al. have noted a strong correlation between pulse splitting and spectral broadening, especially towards lower frequencies [163]. In this case, we remark that the appearance of pulse splitting in the nonlinear-focusing regime also coincides with the observed strong spectral broadening in the lower frequencies for filamentation at 800 nm in air, supporting the universality of the physics governing filamentation for different states of matter.

4.1.4 - Verifying the Generality of the Transition between Regimes

Up to this point, the only condition that has been changed is the geometrical focal distance. To verify that the proposed analytical method to determine the transition between the linear-focusing and nonlinear-focusing regimes is universally applicable, we have to test its validity for a range of initial conditions. To facilitate the comparison of the transition point between different initial conditions, we will define NA_T as the NA when the transition between regimes takes place. Two supplementary results are presented here to demonstrate that the proposed transition is indeed accurate for different initial conditions.
Figure 33. Spectral broadening measurements with 16 mJ pulses in the (a) linear-focusing regime and (b) nonlinear-focusing regime. Geometrical focal distances are indicated in the legends, and the initial beam parameters are identical to those in Figure 31c,d.

The first supplementary result is the measurement of spectral broadening for higher energy pulses shown in Figure 33. With the increase in pulse energy from 3 mJ to 16 mJ, \( NA_T \) was calculated to change from \( 4.28 \times 10^{-3} \) (\( f = 0.99 \) m) to \( 4.01 \times 10^{-3} \) (\( f = 1.06 \) m). Spectral broadening was significantly more pronounced with higher energy, but the defining spectral characteristic between the regimes remained unchanged – spectral broadening in the IR was practically absent in the linear-focusing regime, and quickly became significant in the nonlinear-focusing regime. Spectral broadening was also minimal close to \( NA_T \), and increased for larger or smaller NA values.
Figure 34. Simulated spectral broadening with a smaller initial beam size of \( w(0) = 2.83 \text{ mm} \) in the (a) linear-focusing regime and (b) nonlinear-focusing regime. Geometrical focal distances are indicated in the legends, and their initial beam parameters are identical to those in Figure 31a,b. Inset: Evolution of the on-axis temporal profile of the pulses for \( f = 1 \text{ m} \) and \( f = 2 \text{ m} \).

The second supplementary result is the simulated spectral broadening for a smaller initial beam shown in Figure 34. With the reduction in the initial beam size from \( w(0) = 4.25 \text{ mm} \) to \( w(0) = 2.83 \text{ mm} \), \( \text{NA}_T \) was calculated to change from \( 4.28 \times 10^{-3} \) (\( f = 0.99 \text{ m} \)) to \( 4.25 \times 10^{-3} \) (\( f = 0.67 \text{ m} \)). In Figure 34a, the spectra for \( f = 0.4 \text{ m} \) and \( 0.6 \text{ m} \) behaved as expected, having minimal broadening in the IR. \( f = 0.8 \text{ m} \) belonged in the nonlinear-focusing regime, and slight broadening in the IR began to appear. In Figure 34b, broadening in the IR continued to increase by a small amount for \( f = 1 \text{ m} \), and only became significant for \( f = 2 \text{ m} \). To understand the weak broadening in the IR when the conditions first transitioned from linear-focusing to nonlinear-focusing regime, we turned to the temporal profiles of the filamenting pulses (Figure 34 inset). At \( f = 1 \text{ m} \), pulse splitting was just beginning to manifest, and when the focal distance increased to \( 2 \text{ m} \), pulse splitting was clearly present. This correlation between pulse splitting and spectral broadening strengthens the observations and deductions made in Chapter 4.1.3. The transition as defined by \( \text{NA}_T \) remains accurate, even though the significance of the Kerr effect in the
nonlinear-focusing regime may not be obvious when the geometrical focusing conditions are close to $N_A T$.

Figure 35. Plot of (a) $N_A T$ for a range of initial beam sizes, pulse durations and peak powers. Surfaces in red, blue, brown and gray represent the calculated $N_A T$ values for FWHM beam diameters of 2 mm, 5 mm, 10 mm and 20 mm respectively. (b) shows a cross section of the surfaces for pulse duration of 100 fs and (c) a cross section of the surface for peak power of $10 P_{cr}$.

We calculated the values of $NAT$ for a range of initial beam sizes, pulse durations and peak power, and the results are shown in Figure 35. Within the range of initial parameters considered, which are representative of typical experimental conditions, the variation of NAT is relatively small, between 0.003 and 0.005. This implies that the change in the balance of the physical processes responsible for filamentation is primarily determined by the geometrical focusing condition, and the influence by other parameters such as pulse duration and peak power (or pulse energy) is much less significant.
In fact, for longer pulses (>100 fs) the effects of KSF should be stronger as the Raman Kerr effect can exert an effect on the pulses. This has not been taken into consideration when determining $N_{AT}$. $|s_R|$ would increase more rapidly with propagation if the overall KSF is stronger, leading to effectively higher $N_{AT}$ values for longer pulses and therefore a smaller variation in the overall range of $N_{AT}$ values.

4.1.5 - Discussion

Due to space constrains, as well as to achieve better stability in the filaments, filamentation experiments in the laboratory are often carried out using focusing optics of moderate focal lengths. These conditions are therefore very different from the beams used to generate filaments at long distances, which are often the goal for many applications. Knowledge of the transition between the linear-focusing and nonlinear-focusing regimes is important to ensure that the results derived from filaments in the laboratory are applicable to the filaments out in the field. For example, the contribution of the Kerr effect, and consequently the amount of spectral broadening, will be underestimated in the laboratory if the initial NA is too high. Such laboratory results would then be unsuitable for determining the capabilities of filaments in a LIDAR application.
It is instructive to review the results published by other research groups in the literature, and determine if the observations that have been made are consistent with the linear- focusing and nonlinear-focusing regimes as defined here. The findings are summarized in Table 2. The results of Geints et al. and Talebpour et al. are especially interesting, as a range of conditions covering both regimes were treated in their work. Geints et al. observed sharp changes in the trends for filament length and plasma density for focusing conditions in the vicinity of NA①. Talebpour et al. reported the observation of observation of refocusing
events, which is a behavior of filamentation that is explained by KSF. From their observation, these refocusing events occur only when \( \text{NA} \leq \text{NA}_T \), i.e. in the nonlinear-focusing regime. Some examples of filamentation in argon have also been examined, and the observations reported match the regimes in which the beams have been determined to belong to. This demonstrates that the transition between filamentation regimes is applicable to gases other than air.

It may be important to note that the calculation of \( s_K \) and \( s_p \) requires that the beam maintain a profile comparable to that of a focusing Gaussian beam. This condition is fulfilled for focusing condition in the proximity of \( \text{NA}_T \) and larger, but in low-NA beams KSF will modify the beam convergence significantly prior to the geometrical focus. For very high NA conditions, other phenomena such as small-scale filamentation and “superfilamentation” will take place and the description of the linear-focusing regime may no longer be accurate. Since the only processes considered in this analysis are focusing and defocusing effects, the validity of this analysis may not extend to cases where other effects such as dispersion and absorption become significant. This could apply to, for example, the filamentation of few-cycle pulses with extremely broad spectra and filamentation in condensed media.

4.1.6 - Summary

In summary, we have described an analytical method to distinguish between two regimes of filaments generated in gases based on the initial geometrical focusing condition. In the
high-NA or linear-focusing regime, KSF has been shown to play a significantly less important role in filamentation than for filaments in the low-NA or nonlinear-focusing regime. As a result, the properties of a filament in the linear-focusing regime differ noticeably from that of a “typical” filament. For filamentation in air at 800 nm, the transition focusing condition between the linear-focusing and nonlinear-focusing regimes fall within the range $NA_T = 0.003$ to $0.005$ for common experimental conditions. A review of results published in the literature reveals that the transition between the regimes accurately describes observations that had been made in multiple independent experiments.

4.2 - Molecular Effect on Elliptically Polarized Filaments

Other than the geometric focusing condition, another initial condition that an experimenter or laser user could easily modify is the polarization of the beam. As air (and gases in general) is an isotropic medium, the rotation of a linearly polarized beam is expected to have no effect on the propagation; modifying the polarization from linear to elliptical and to circular, on the other hand, could change the way the beam interacts with the medium and therefore change the properties of the filaments formed.

Several research groups have examined the effects of different polarization on filamentation. Certain effects of polarization on filamentation are generally accepted. For example, Fibich and Ilan showed that the critical power for self-focusing of a circularly polarized beam (CP) is greater than that of a linearly polarized one (LP) [167]; Ammosov et
al. demonstrated that photoionization is less efficient for CP than for LP in argon [168]; CP has also been shown to be more resistant to breaking up into multiple filaments than LP in the right conditions [169, 170]. However, contradicting results from studies of CP filaments have also been reported. While it appears intuitive that weaker Kerr effect and ionization efficiency in CP should lead to less efficient supercontinuum generation [159, 171, 172] and lower plasma densities [30], results from various groups have also shown more efficient supercontinuum generation [173, 174] and higher plasma densities from CP filaments [175]. Moreover, there is still some uncertainty over the stability of elliptical polarization when the pulse undergoes filamentation [30, 167, 176–178].

We have observed an anomalous spectral broadening behavior at elliptical polarizations near CP for filaments in air. To better understand the anomalous spectral broadening, we examined spectral broadening in pure molecular and atomic gases for different initial polarizations, and also measured the ellipticity of the beam after filamentation.
4.2.1 - Experimental Setup

The experimental setup used to study polarization effects in different gases is shown in Figure 36. The central portion of the gas tube was made of transparent PVC, and the ends of the tube were sealed by fused silica windows (800 nm AR coated for entry and uncoated for exit) that allowed the tube to be pumped down and filled with gases up to pressures of ~1 atm. The pulse energy and duration were fixed at 2.8 mJ and 50 fs respectively, and the pulse polarization was controlled by a zero order quarter-waveplate (CVI QWPO-800-10-4). The pulse was then focused by a \( f = 3 \) m lens and the filament formed near the middle of the gas tube.

To measure the ellipticity of the beam after the tube, its energy was attenuated by an ND filter before it was sent through a broadband polarizing beam cube. The cube was rotated along the propagation axis, and the energy transmitted through the cube was measured by
an energy meter (Gentec QE25LP) for different polarizing cube angle. The amplitude of the modulation in the transmitted energy could then be used to deduce the ellipticity of the beam. To measure the spectrum of the supercontinuum generated, a white diffusing screen was inserted after the tube, and a multi-mode fiber was used to collect the diffused light into a spectrometer (Ocean Optics HR2000 for visible light, USB2000 for NIR). To determine the position and length of the filaments, pictures of the filaments were taken through the gas tube using a DSLR (Canon 5D mk II).

Some precautions were taken to ensure that the observations were comparable across the different gases. For all the gases, the pressures were kept low enough such that multiple filaments were not formed (visual verification by observing the supercontinuum pattern) for any polarization. The exit window was kept far away enough from the filament to ensure that nonlinear effects in the window were minimal, if not completely eliminated. Both spectral and ellipticity measurements were made on the central core of the beam; the conical emission (colored rings around the core) was removed by irises in the ellipticity measurements, and rejected by the acceptance angle of the collection fiber in the spectral measurements. For certain conditions, filamentation restarted in air after exiting the tube, due to self-focusing in the exit window, or bright supercontinuum generation in the window could be observed; both of these situations were avoided for the measurements.
4.2.2 - Measurements and Results

4.2.2.1 - Spectral Broadening

Figure 37. Spectral measurements for different initial polarizations in (a) vacuum and in (b) air. The color-map follows a logarithmic scale for better visibility of the broadened spectra. $0^\circ$ and $90^\circ$ positions of the QWP correspond to an initial linear polarization of the pulse, while $45^\circ$ correspond to circular polarization.

To ensure that there were no undesired effects causing any changes in the spectrum in the experimental setup, the spectrum of the pulse was first recorded for different initial polarizations with the gas tube pumped down to a vacuum ($< 0.1$ mTorr). Figure 37a shows that changing the initial polarization of the pulse from linear through elliptical to circular has no effect on the output spectrum. This spectrum was verified to match that of the pulse before entering the gas tube.

When the tube was filled with air, the spectrum at the output of the tube was perceived by eye to broaden significantly in LP (QWP angle $0^\circ$), as the beam became bright red and highly visible. As the polarization was changed from LP to elliptical, the visibility of the beam decreased gradually, with minimal visibility reached with a QWP angle of $\sim 35^\circ$. However, when the QWP was rotated from $35^\circ$ to $39^\circ$, the visibility of the beam increased.
rapidly and acquired an orange hue. This anomalous spectral broadening behavior reversed when the QWP was rotate from 39° to 43°, and the beam reached its dimmest point at CP (QWP angle 45°). The spectral broadening pattern was symmetrical as we continued to rotate the QWP from 45° to 90°. The spectral evolution displayed in Figure 37b accurately reflects the trend observed by eye in the visible wavelengths, and shows that a similar broadening trend occurs towards longer wavelengths as well.

To help deduce the origins of the anomalous broadening, we examine the spectral broadening behavior with changes in polarization in different molecular and atomic gases.
Figure 38. Spectral measurements for different initial polarizations in (a) nitrogen, (b) oxygen, (c) argon and (d) krypton.

Figure 39. Images of the beam at the output of the gas tube when filled with 0.7 atm of nitrogen (top) and argon (bottom), for different QWP angles changing the polarization from linear to circular.
In nitrogen (Figure 38a), very similar behavior to air was observed. The anomalous broadening at near-circular polarization was significantly more pronounced, producing a white core at the center of the beam at QWP angles of $39^\circ$ and $51^\circ$ (Figure 39). The anomalous broadening effect was observed in oxygen as well (Figure 38b), but this time it was weaker than in air. The QWP angles at which anomalous broadening peaked were also different - $43^\circ$ and $47^\circ$. In argon and krypton (Figure 38c,d), anomalous broadening clearly did not occur. Instead, the spectrum at LP was significantly broader than in the molecular gases, and the spectrum in the visible shrunk gradually and smoothly as the polarization was changed to CP. This effect was easily visible on the diffusing screen (Figure 39). The spectrum in the NIR, on the other hand, was relatively unchanged by rotation of the QWP.

4.2.2.2 - Ellipticity

![Ellipticity measurements of the beam after filamentation through different gases.](image)

The ellipticity measurements of the beam after filamentation exhibited a similar distinction between molecular and atomic gases (Figure 40). For better perspective, the ellipticity...
measurements were compared to that of the beam propagating through vacuum (plotted in gray). In nitrogen and oxygen, the beam exhibited slightly higher ellipticity than in vacuum as the polarization was changed from LP. At the QWP angles where anomalous spectral broadening occurred, the output ellipticity also showed a significant decrease to values below that in vacuum. The ellipticity increased again towards CP, but not to the same value as the vacuum case. In atomic gases, the ellipticity increased more rapidly when the polarization was changed from LP, and reached a plateau at QWP ~30° with a value of ~0.95.

It is important to note that the measurements made cannot distinguish between circular/elliptical polarization and depolarization. Nonetheless, depolarization was not expected to occur in vacuum, therefore the vacuum data would remain as a valid point of reference for the other measurements. In addition, a decrease in ellipticity, which is observed in the molecular gases, is a clear indication that the output polarization is closer to LP and is not affected by the CP or depolarization ambiguity.
4.2.2.3 - Filament Position and Length

Figure 41. Images of the filament in 0.7 atm N₂ for different QWP angles (top) and the corresponding map of plasma emission (bottom, in linear scale). The aspect ratio of the images has been altered to increase visibility of the filaments, they look much narrower in the original images.

As nitrogen exhibited the most pronounced anomalous spectral broadening, images or the filament were taken to examine the correlation between that and the filament’s length and position. The images (integrated over 300 shots) are shown in Figure 41, and plasma emission data was extracted from the images to construct a corresponding plasma emission map. From the images and the map, the filament can be clearly observed to have begun further from the lens as the polarization was changed from LP to CP. This trend was
maintained even at the anomalous broadening QWP angle. However, the filament was instead lengthened away from the lens when anomalous broadening occurred.

4.2.3 - Discussion

The spectral broadening and ellipticity measurements show that anomalous behavior occurs only in molecular gases. Therefore, to understand the origin of the anomalous behavior, we look for polarization related processes that are present only in molecules. If the effect(s) responsible for the anomalous behavior manifests during the self-focusing phase of the beam propagation, it would likely have an impact on the starting position of the filament. Based on the observations in Figure 41, that was not the case and the filament was instead lengthened towards the end. We can therefore deduce that the process(es) responsible for the anomalous behavior only takes place in the filament, under the effect of high optical fields and intensities. It could also be an ionization related process.

Palastro suggested that the coupling between polarizations via molecular rotation effects has been under-estimated in the simulations of filamentation [177]. By including the off-diagonal terms in the nonlinear susceptibility tensor for molecular rotation, he observed stronger exchange of energy over the course of filamentation between orthogonally polarized fields. More importantly, he also obtained an unusual increase in the degree of polarization and the ellipticity of the post-filamentation beam near CP. Despite the difference in the sign of the change in ellipticity between our experimental results and Palastro’s simulations, it is highly probable that the results are correlated. Filaments
created by a near CP beam may therefore be prolonged and spectrum broadening increased due to the exchange of energy between polarization components.

In an attempt to verify if increasing the impact of molecular alignment on filamentation can significantly increase the anomalous spectral broadening and ellipticity behavior, the experiment was repeated with the pulse positively chirped to produce an initial pulse duration of 150 fs, and the initial pulse energy increased to 8.3 mJ to maintain a similar peak power to critical power ratio. However, in the chirped pulse condition it was difficult to avoid multi-filamentation (see Chapter 4.2.3.2), and the spectral broadening was generally weaker and more irregular. The measurement was abandoned as the observations were deemed to be inconclusive.

In an experiment to study the effects of polarization on the backwards-lasing phenomenon in filaments in nitrogen, Mitryukovskiy et al. [179] have observed that CP and near circular polarization produced the highest gain in backwards stimulated emission in a filament in nitrogen. They attributed the results to the high kinetic energies of the free electrons in the CP case. Varela et al. [170] explained the distribution of multiple filaments in nitrogen by the anisotropic electronic density in a filamenting beam due to the orbital shape of the molecule. Both of these effects could contribute to the anomalous behaviors observed in our experiment. The decrease in ellipticity at the QWP angles where anomalous broadening occurred suggests that the electric field component polarized along the minor axis suffers
from higher losses, due to either anisotropy in the photoionization process or subsequent plasma absorption, or a combination of both.

The observation in air (Figure 37b) resembles that of nitrogen (Figure 38a), except that the anomalous broadening effect is weaker. This is similar to the observations of Mitryukovskiy et al. [179], who noted that the lasing effect in the nitrogen filament diminished rapidly when oxygen was added.

4.2.3.1 - Simulations with Cross-Kerr Effect

In another attempt to determine the contribution of molecular alignment effects to the observed anomalous behavior, the NLSE code described in Chapter 2.2 was modified to account for the polarization of the pulse. The modification was made according to the formalism described by Kolesik et al. [30] and Couairon et al. [19]. The left-circular and right-circular polarized electric fields, $\varepsilon^-$ and $\varepsilon^+$, are coupled in the NLSE by the cross-Kerr effect (both instantaneous and Raman Kerr). Compared to Equation (9), the polarization-coupled NLSE is therefore given by
\[
\frac{\partial \varepsilon^\pm}{\partial z} = \frac{i}{2k} \nabla_\perp^2 \varepsilon^\pm - \frac{k''}{2} \frac{\partial^2 \varepsilon^\pm}{\partial t^2} + i \frac{2 k_0 n_2}{3} (1 - f) \left( |\varepsilon^\pm|^2 + 2 |\varepsilon^\mp|^2 \right) \varepsilon^\pm \\
+ i \frac{2 k_0 n_2}{3} \int_{-\infty}^{t} R(t - t') \left( |\varepsilon^\pm(t')|^2 \right) dt' \varepsilon^\pm - \frac{\sigma}{2} (1 + i \omega_0 \tau_c) \rho \varepsilon^\pm \\
- \frac{\beta}{2} |\varepsilon|^{2K-2} \left( 1 - \frac{\rho}{\rho_{nt}} \right) \varepsilon^\pm
\]

Equation (27)

The total intensity is given by \( |\varepsilon|^2 = |\varepsilon^-|^2 + |\varepsilon^+|^2 \). The ionization rate equation (Equation (6)) is not modified as the photoionization process is assumed to be independent of the field polarization in this case.

Figure 42. Simulated spectra for different initial polarization, with cross-Kerr effect (both instantaneous and Raman) taken into consideration.

The simulation was carried out in air for different initial polarization as defined by the QWP angle. The initial pulse duration and energy in the simulation were 50 fs and 0.8 mJ,
and the beam was focused at a distance of 3 m. The results in Figure 42 show a behavior that is more akin to the atomic gases (Figure 38c,d), with a smooth decrease in the spectral broadening when changing from LP to CP. The trend in Figure 42 is also consistent with the results of Kolesik et al. [30], where they showed weaker spectral broadening in CP compared to LP in their simulations.

It is therefore clear that a simple modification of the NLSE to include the cross-Kerr effect, even with molecular alignment effects accounted for in the Raman Kerr term, is insufficient to account of the complex dynamics taking place for elliptically polarized filaments in molecular gases. Varela et al. [170] made a similar remark as their simulations failed to replicate some of the trends in their experimental observations with elliptically polarized and CP filaments. A more detailed model of the rotational dynamics of the molecules need to be taken into account, as Palastro has done [177], before the full impact of polarization on filamentation can be realized in a numerical study.

An additional remark should be made regarding the simulation results of Kolesik et al. [30] and the ellipticity measurements in Figure 40. It is possible that a simple addition of the cross-Kerr effect to the NLSE is effective for describing filaments of arbitrary polarization in atomic gases. Kolesik et al. noticed a strong depolarization effect of filamentation on an initially elliptically polarized beam. This depolarization did not occur for beams that were initially LP or CP, or close to one of these polarizations. This could account for the apparent
increase in ellipticity and plateau in Figure 40 for argon and krypton, since the measurement did not distinguish between CP and depolarization.

4.2.3.2 - Effects of Multi-filamentation

Figure 43. Spectral measurements and images of the beam for cases where multi-filamentation took place.

When the pressure of the gases were too high, multiple filaments began to form in the tube. In the case of nitrogen at 1 atm (Figure 43a,c), we observed two distinct filaments, and the anomalous spectral broadening occurred at two different sets of QWP angles. Upon closer inspection, it can be deduced that each of the filament exhibit anomalous spectral broadening at a different QWP angle; even in the same gas, the abrupt increase in spectral broadening efficiency can occur at different QWP angle depending on the initial conditions.
This outlines the importance of ensuring that only a single filament was formed during the experiments as the contribution of individual filaments could be difficult to isolate.

In 1 atm argon, multi-filamentation was also observed (Figure 43b,d) for polarization closer to LP. Compared to the results of at 0.7 atm, the spectrum at 1 atm exhibited very weak dependence on initial polarization. As the QWP was rotated, the distribution of filaments could be observed to change, and at least one bright white core was always present. The break up of the beam into multiple filaments for polarization closer to LP could be limiting the amount of spectral broadening generated, as energy is divided and channeled into the individual filaments. This reduction in overall spectral broadening due to multi-filamentation therefore makes the observation of polarization effects similar to that in Figure 38c impossible.

4.2.3.3 - Effects of Focal Distance

On a separate occasion, the spectral measurements for different QWP angles was repeated in air without the gas tube, and for lenses of different focal lengths (f = 0.2 m, 0.4 m, 0.6 m, 0.7 m, 1 m, 2 m, 3 m and 5 m). The results that best represent the characteristics observed are shown in Figure 44. The pulse duration and energy were 58 fs and 4.5 mJ respectively.
Figure 44. Spectral measurements in 1 atm air using lenses of different focal lengths.

For focusing conditions in the linear-focusing regime (see Chapter 4.1), represented by $f = 0.4$ m (Figure 44a), the trend in air is similar to what has already been observed for atomic gases (Figure 38c,d). At the transition point between linear-focusing and nonlinear-focusing regimes (Figure 44b), spectral broadening is minimal, and the effects of polarization are barely noticeable. Compared to the spectrum at LP, the spectrum at CP carried slightly more energy in the shorter wavelengths and less energy in the longer wavelengths, though the spectral width remained comparable.
Anomalous spectral broadening only manifested in the nonlinear-focusing regime (Figure 44c,d). The spectrum in Figure 44c is similar to those previously shown in the same focusing condition. The oscillations in the spectrum in this case could be due to stronger pulse splitting or interference between multiple filaments, or both. With a longer focal distance (Figure 44d), and therefore longer filaments, anomalous spectral broadening occurred at multiple sets of QWP angles (32°, 40° and 45°) due to multi-filamentation. The difference in the QWP angles where anomalous broadening occurs between the spectra in Figure 44c and Figure 44d demonstrates that the initial polarization state that can lead to anomalous broadening likely depends on the filament length. In the right conditions, the spectral broadening at CP can be comparable or even greater than that at LP. This can explain the results demonstrated by Yang et al., where they observed more efficient spectral broadening with CP [174].

4.2.4 - Summary

We have presented measurements that exhibit anomalous spectral broadening behavior for filamenting pulses with specific initial polarization ellipticity near CP. This anomalous behavior is only observed in molecular gases, and leads to a decrease in the final ellipticity of the beam as well as an increase in the length of the filament. The mechanism responsible for this effect has yet to be identified, though it is clear that it is only active after filamentation has commenced. Coupling between polarization components due to nonlinearity from molecular rotation may be responsible for the anomalous behavior, but a more in-depth investigation will need to be conducted before it can be verified.
These results demonstrate the strong dependence of the filamentation process on the initial polarization of the beam, and how the outcome of filamentation can be affected. Polarization control may therefore be an important addition to applications of filamentation where the generation of supercontinuum is a key consideration, such as white light LIDAR and the generation of few-cycle pulses.

4.3 - UV Filaments in Air

Even though the Ti:sapphire laser is the laser system used in the majority of filamentation studies, there are efforts to extend the investigation and application of filaments to UV wavelengths as well. UV filaments have been studied for various applications, including guiding electrical discharges [180, 181], inducing condensation [53], and LIBS [79, 182]. There are several advantages to working with UV pulses as opposed to NIR ones. The critical power for self-focusing is an order of magnitude smaller, therefore less energy is required for filamentation to occur. The photons carry more energy, and can interact more strongly with the propagation medium or targets that the filament encounters. In certain cases, UV filaments have also been shown to carry more energy in its core [140] and generate more free electrons in its plasma channel [183].
Table 3. Experimentally determined characteristics of femtosecond UV (248 nm) filaments.

<table>
<thead>
<tr>
<th>Laser parameters</th>
<th>Schwarz et al. [29]</th>
<th>Tzortzakis et al. [183, 184]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pulse duration</td>
<td>1.1 ps, positively chirped from 600 fs</td>
<td>450 fs</td>
</tr>
<tr>
<td>Energy</td>
<td>up to 25 mJ</td>
<td>2 mJ and 5 mJ</td>
</tr>
<tr>
<td>Focusing condition</td>
<td>1 cm diameter, collimated</td>
<td>7 mm diameter, 9.5 m focusing</td>
</tr>
<tr>
<td>Intensity</td>
<td>1-2 TW/cm²</td>
<td>~0.1 TW/cm²</td>
</tr>
<tr>
<td>Plasma density</td>
<td>3x10^{15} cm⁻³</td>
<td>10^{16} cm⁻³</td>
</tr>
<tr>
<td>Filament size</td>
<td>120±20 μm</td>
<td>150 μm</td>
</tr>
<tr>
<td>Spectral broadening</td>
<td>negligible</td>
<td>Only towards longer wavelengths</td>
</tr>
</tbody>
</table>

Despite the potential benefits, very few fundamental studies have been conducted on the properties of UV filaments. Some of the findings of two notable experimental investigations into the properties of femtosecond UV filaments have been summarized in Table 3. Some other reported observations of UV filaments, such as Chalus et al. [140] and Dergachev et al. [185], are either in the picosecond regime (and therefore may involve very different dynamics and physical processes) or lacking in detailed experimental results.

Numerical and theoretical studies of UV filaments are more frequently encountered in literature. Couairon and Bergé reported numerical results of filaments at 248 nm with significantly higher intensities (up to 4 x 10^{13} W/cm²) and plasma densities (above 10^{17} cm⁻³) [18]. They explained that the discrepancy was normal as experimental results only retrieve average quantities, which they calculated to be significantly lower in the case of UV filaments. Skupin and Bergé examined the spectral broadening trend with different...
central wavelengths, and found that supercontinuum generation weakens with shorter wavelengths [186]. They explained that shorter wavelength pulses experience greater losses, therefore the filaments are shorter and produce a weaker supercontinuum. They also showed that shorter UV pulses will generate broader supercontinua, due to the higher maximum intensities reached. Fedorov and Kandidov theoretically and numerically studied filament properties for wavelengths ranging from 248 nm to 1.24 μm, and found that UV filaments have lower intensity and are smaller in size, but have higher plasma densities [187]. Due to the low intensity threshold of ionization in the UV, the low intensity portions of the UV beam would be sufficient to lead to a low level of ionization, creating a weak plasma pedestal around the filament core [188]. Schwarz and Diels predicted that due to the $I\lambda^2$ dependence of the rate of energy loss to inverse bremsstrahlung, it would be possible to create kilometer-scale filaments with nanosecond UV pulses [143, 189]. However, Niday et al. demonstrated that such a long pulse should be unstable as a filament, and would fragment in time and rapidly diffuse [190].

In the experimental characterization of UV filaments highlighted in Table 3, relatively long pulses were used. Between the two results in Table 3, there are both quantitative and qualitative differences, probably due to the different pulse durations. With ultrafast sources available now in the sub-100 fs range, it would be instructive to revisit the properties of UV filaments with these pulses. Short pulses in the UV can be obtained by direct third harmonic generation (THG) of the output of a Ti:sapphire laser. Moreover, to obtain the power required for filamentation with the longer pulses, the output of either a
Ti:sapphire (frequency-tripled) or dye laser (frequency-double) had to be further amplified. Shorter pulses do not need the same level of energy to reach the power necessary for filamentation. Not requiring an extra amplification stage would make UV filaments more accessible to many laboratories, and would make switching between NIR and UV sources for an experiment a simpler task.

4.3.1 - Third Harmonic Generation of an Ultrafast Pulse

In principle, the third harmonic of the 800 nm Ti:sapphire pulse can be generated in a single step via a $\chi^{(3)}$ process in a nonlinear medium. However, the conversion efficiency of such a process is very low and phase matching is difficult to achieve. Direct conversion of the fundamental to $3\omega$ is therefore not suitable for generating high energy pulses in the UV. To achieve higher conversion efficiency, a cascaded $\chi^{(2)}$ process is often used instead. The 800 nm fundamental pulse first undergoes a frequency doubling process to have part of its energy converted to 400 nm. The fundamental and $2\omega$ pulses are then mixed in a second nonlinear crystal to generate a $3\omega$ pulse via sum-frequency generation.
The schematic of the THG setup used is shown in Figure 45. The key components are, in the order which the beam encounters them, the second harmonic generation (SHG) BBO crystal, group velocity delay (GVD) compensation crystal, dual-wavelength waveplate, and the THG BBO crystal. The fundamental pulse is first propagated through a 15×15×0.2 mm BBO crystal (Quantum Technologies) cut at $\theta = 29.2^\circ$ for Type I SHG. Before the fundamental and $2\omega$ pulses can be mixed in the THG BBO crystal, the difference in their polarization and group delay have to be corrected. As the $2\omega$ pulse generated is polarized perpendicularly to the fundamental pulse, a zero-order dual-wavelength waveplate (EKSMA Optics 466-4211, $\lambda/2$ @ 800 nm and $\lambda$ @ 400 nm) is needed to rotate the fundamental polarization to match that of the $2\omega$. In addition, due to the difference in the group velocities of the fundamental and $2\omega$ pulses in the SHG BBO crystal and dual-wavelength waveplate (air contributes as well, but the effect is negligible), the $2\omega$ pulse would experience greater delay with respect to the fundamental pulse. The GVD compensation crystal (EKSMA Optics 225-2113) is a birefringent calcite crystal with its slow axis aligned to impart greater delay to the fundamental pulse than the $2\omega$ pulse, such
that they would be synchronized upon arrival at the THG BBO crystal. The $3\omega$ pulse is then generated in a 15x15x0.1 mm BBO crystal (OptoCity) cut at $\theta = 44.3^\circ$ for Type I sum-frequency generation. To separate the $3\omega$ pulse from the other pulses, a pair of dichroic mirrors (EKSMA Optics 042-2485, HR @ 267 nm and HT @ 400+800 nm) is used.

4.3.1.1 - Simulation of the Third Harmonic Generation Process
For the purpose of verifying the correct performance of the THG setup, a simulation of the THG process was performed. The equations used to describe the SHG and sum-frequency generation were as described by Boyd [191]. To account for dispersion, a term describing the dispersion of the BBO crystals was added to each of the equations. Using the same system of notation as in Chapter 1.1, the SHG process in the first BBO crystal can therefore be described by

$$\frac{d\varepsilon_1}{dz} = \frac{i\omega_{1}^{2}d_{eff}}{n_{0,1}\varepsilon_{0}k_{1}c^{3}}\varepsilon_{2}\varepsilon_{1}^{*}e^{-i\Delta k z} - i\frac{k_{1}''}{2}\frac{\partial^{2}\varepsilon_{1}}{\partial t^{2}}$$

$$\frac{d\varepsilon_2}{dz} = \frac{i\omega_{2}^{2}d_{eff}}{2n_{0,2}\varepsilon_{0}k_{2}c^{3}}\varepsilon_{1}^{2}\varepsilon_{2}^{*}e^{i\Delta k z} - i\frac{k_{2}''}{2}\frac{\partial^{2}\varepsilon_{2}}{\partial t^{2}}$$

$$\Delta k = 2k_{1} - k_{2}$$

Here, the subscripts 1 refer to the fundamental frequency pulse and subscripts 2 refer to the second harmonic pulse, and $d_{eff}$ is the effective nonlinear tensor coefficient of the process. Similarly, for the THG process in the second BBO crystal,

$$\frac{d\varepsilon_1}{dz} = \frac{i\omega_{1}^{2}d_{eff}}{n_{0,1}\varepsilon_{0}k_{1}c^{3}}\varepsilon_{3}\varepsilon_{2}^{*}e^{-i\Delta k z} - i\frac{k_{1}''}{2}\frac{\partial^{2}\varepsilon_{1}}{\partial t^{2}}$$
The subscripts 3 refer to the third harmonic pulse, and $d_{eff}$ here is different from in the SHG case as the axes of the BBO crystals are not oriented in the same directions.

To numerically solve the coupled equations given above, the split-step Fourier method as described in Chapter 2.2.1 was used. The nonlinear terms were resolved in the temporal domain and the dispersion term in the spectral domain. Diffraction was not taken into account, therefore it was not necessary to carry out calculations in the spatial frequency domain, even though the calculations were fully resolved in both time ($t$) and space ($r$). In addition, to account for GVD, a relative delay between the pulses based on the group velocity of each pulse was introduced after each simulation step.

The simulation step size was fixed at 1 μm, and the refractive index, dispersion and group velocity values at each wavelength were calculated based on the Sellmeier equation of BBO [192]. The GVD effects of the GVD compensation crystal and the dual-wavelength waveplate was assumed to be a nett shift of the $2\omega$ pulse forward by 40 fs (the relative delay between the fundamental and $2\omega$ pulses after the SHG BBO crystal from simulation results), such that the two pulses were overlapped in time upon arrival at the THG BBO.
\( d_{\text{eff}} \) was used as a fitting parameter (for both BBO crystals) to attempt to match the measured results. As an initial estimate, the values provided by Klein et al. were used:

\[
d_{\text{eff}} = d_{31} \sin \theta - d_{22} \cos \theta \sin 3\varphi
\]

(35)

where \( d_{31} = 0.26 \text{ pm/V} \), \( d_{22} = 2.11 \text{ pm/V} \) and \( \varphi = 90^\circ \).

4.3.1.2 - Third Harmonic Generation Results

![Figure 46. Simulated and measured energies of the fundamental and second harmonic pulses after the first BBO crystal.](image)

Before assembling the full THG setup, the energies of the fundamental and \( 2\omega \) pulses after the first BBO crystal was measured with an energy meter (Gentec QE25LP) and compared to the simulated results. The pulses were separated by a pair of dichroic mirrors for the measurement. The measured values showed good agreement with the simulated values. The input fundamental pulse was assumed to have an intensity FWHM of 5 mm and a pulse duration of 50 fs.
Figure 47. Simulation and measured results of THG. (a) shows the simulated and measured pulse energies at the different wavelengths, (b) shows the simulated temporal profile of the \(3\omega\) pulse for an input fundamental pulse energy of 3 mJ, and (c) shows the measured beam profile of the \(3\omega\) beam, also with 3 mJ input.

With the THG setup fully assembled, the final pulse energies at the three different wavelengths were measured and compared to the simulated results (Figure 47a). For the \(3\omega\) beam, two measurements were taken at each energy. The first is a measurement directly after the pair of dichroic mirror, which consist of the \(3\omega\) pulse energy as well as any residual fundamental and \(2\omega\) energy. For the second measurement, a thin sheet of borosilicate glass (Fisherbrand microscope cover glass 12-544G, 0.16-0.19 mm thickness) was inserted just before the energy meter to absorb all the \(3\omega\) energy while transmitting most of the residual fundamental and \(2\omega\) energy. The actual \(3\omega\) energy was taken to be the difference of the two measurements. The residual fundamental and \(2\omega\) energy was relatively small at high energies (~2% of ~500 µJ total measured energy), but became significant at low energies (~20% of ~20 µJ). A calibration curve was deduced from these
measurements for use in subsequent experiments. The measured and simulated values showed good agreement at lower energies, but began to differ at higher energies. These differences could be due to the spatial and temporal profiles of the laser output not being perfectly Gaussian. The differences are greater at high energies as the sensitivity of the cascaded nonlinear process increases with the intensities involved. The accuracy of the simulation can be considered to be good for input energy up to 3 mJ, or $3\omega$ output $\leq 500 \, \mu\text{J}$. This was also the highest energy used in subsequent experiments, as we were attempting to characterize single filaments and multi-filamentation began to occur for $3\omega$ energies greater than 200 $\mu\text{J}$.

The pulse duration of the $3\omega$ pulse was determined from the simulation results to be 56 fs (Figure 47b). The simulation also showed that the pulse was slightly positively chirped, which was consistent with the propagation through the various normally dispersive crystals. The beam profile was measured based on the fluorescence generated by UV excitation of a borosilicate glass sheet, as the filters in front of standard CCD sensors block UV emission below 300 nm making direct imaging of the beam impossible. The fluorescence was determined to increase linearly for the range of fluence to be measured by the following method. A small central region of the UV beam was selected by an aperture, allowing the beam to be approximated to have a flat-top profile. The fluorescence generated by the flat-top beam was then captured by a CCD camera (The Imaging Source DMK22BUC03 with 35mm f/1.4 C-mount lens) at different pulse energies, and the total pixel values compared to the energy in the flat-top profile. The beam profile at 3 mJ input
energy is shown in Figure 47c. The simulations predicted a slightly smaller beam with 4 mm intensity FWHM.

![Figure 48](image.png)

Figure 48. Beam profile measurements of the $3\omega$ beam around the focus of a 2 m lens. The data retrieved from ten different images per position are represented by the errorbars, and the solid lines represent the fitting of the points by a Gaussian profile.

The $3\omega$ beam was focused using a $f = 2$ m lens, and its beam profile was recorded using the modified grazing incidence imaging system described in Chapter 4.3.2.2. For this measurement, the $3\omega$ energy was reduced to 8 $\mu$J, which was below the threshold for filamentation. The horizontal and vertical sizes of the beam could be well fitted by Gaussian profiles, although some level of astigmatism was observed. The profile of the $3\omega$ beam could therefore be approximated by a Gaussian beam. This is important for subsequent comparison of the filament profiles with that of the linearly propagating beam.
The spectrum of the 3ω output was measured using a fiber coupled spectrometer (Ocean Optics HR2000+). The light was scattered off a ground CaF₂ plate, collected by a fused silica lens into the UV transmitting multi-mode fiber and sent into the spectrometer. The measured and simulated spectra matched well (Figure 49a), although the measurements showed a slight symmetric broadening of the 3ω spectrum with energy, which was not observed in the simulations (Figure 49b,c). The spectra in Figure 49b have been normalized to a peak value of unity to make the gradual broadening of the spectrum with energy more visible.

4.3.2 - Observation of UV Filaments

With the 3ω beam well characterized, we could proceed to create filaments in the UV – in the 3-photon ionization regime of air (considering oxygen as the primary ionizing species [18], and 4-photon ionization of nitrogen) – and observe how the behavior of these filaments compare to those of filaments at 800 nm. For these observations, the 3ω pulse was focused with a fused silica lens of 2 m focal distance. The results could therefore be qualitatively compared to those of the 800 nm pulse previously shown in this dissertation.
(e.g. Figure 6, Figure 12 and Figure 13). The Sellmeier equation of fused silica was used to calculate the effect of dispersion on the pulse, and the pulse was determined to be further positively chirped from 56 fs to 75 fs after transmission through the lens. Energy measurements were carried out after the lens to account for losses from reflections off the lens surface.

Using the focusing regime analysis of Chapter 4.1, the transition between linear-focusing and nonlinear-focusing regimes was found to be \( \text{NA}_r \approx 0.0078 \) to 0.009 for the range of pulse energy under consideration. A focal distance of 2 m therefore meant that filamentation was occurring in the nonlinear-focusing regime. Using \( n_2 = 8 \times 10^{-19} \text{ cm}^2/\text{W} \) [18], the critical power for self-focusing at 267 nm was calculated to be 134 MW. For a pulse duration of 75 fs, this corresponded to a pulse energy of 10.7 \( \mu \text{J} \).
4.3.2.1 - Filament Position and Length

Figure 50. Images of the plasma emission from (a) filaments with the 267 nm pulse and (b) filaments with the 800 nm pulse. The beams were focused with a 2 m lens, and the white dashed lines indicate the positions of the geometrical focus. The Rayleigh zones in each case are also indicated.

As an initial verification of filamentation, the plasma channel created by the filament was observed, and seen to lengthen with increasing pulse energy. Images of the plasma emission were recorded with a DSLR camera (Canon 5D mkII) for different pulse energies, for both the UV and NIR pulses (with the THG setup removed). For the UV pulses, a faint plasma channel was observed for $1.9P_{cr}$, and for peak powers $3.5P_{cr}$ and greater the became progressively brighter and the starting position of the channel moved close to the lens. The rate at which the plasma emission faded away past the focal point was not significantly changed by the pulse energy, which is typical of filaments formed from
geometrically focused beams. In comparison, the plasma channel of the NIR filament exhibited a similar trend, although the absolute length of the channel increased more rapidly with pulse energy / peak power.

![Figure 51](image)

Figure 51. Start position of filaments based of images of plasma emission. Dashed lines represent fitting with Marburger's formula of self-focusing distance.

From the images in Figure 50, the position where filamentation started could be represented by the location where the plasma emission reached half of its maximum value. These positions are plotted in Figure 51. The theoretical collapse distance of the beam was calculated using Marburger's formula (Equations (3) and (4)), and fitted to the filament start positions by allowing the value of the focal distance to be slightly varied from 2 m. The good agreement between the measurements and the theoretical curve for the UV pulse is a confirmation that the estimated pulse duration of 75 fs (which was used to derive the peak power of the pulse, $P_0$) is accurate.
4.3.2.2 - Filament Profile

Figure 52. Schematic of the modified grazing incidence imaging system for imaging UV beams.

The original grazing incidence imaging system could not be used to capture the profile of the UV filaments as the optics used, including the filter in front of the CCD camera sensor, absorbed UV radiation strongly. A modified version of the imaging system, shown in Figure 52, was therefore assembled to capture profiles of the UV beam. The standard CCD camera was replaced by an iCCD camera (Princeton Instruments PI-MAX II) that was sensitive to wavelengths down to 200 nm. As ND filters could not be used, the angles of the wedges were modified to provide sufficient attenuation (~2 × 10⁻⁷) so as to not saturate the iCCD. The imaging lens used was a fused silica lens. The Fresnel reflections off the wedges are highly dependent on polarization. The residual 800 nm and 400 nm light in the beam are S-polarized and therefore subjected to significantly lower attenuation (~0.009). Two bandpass filters (Edmund Optics 67-809) were inserted before the iCCD to reduce the intensity of the residual 800 nm and 400 nm light by >8 orders of magnitude. To verify that the residual light does not interfere with the UV beam profiles, the UV beam was selectively removed by inserting a thin sheet of borosilicate glass before the iCCD, the image background was established to be uniform and identical to that with the beam completely
The magnification of the images on the iCCD (object plane being the first wedge in the imaging system) was determined to be 3.2x.

Using the grazing incidence imaging system, the filament profiles were recorded for three different pulse energies, corresponding to the observations in Figure 50 with the highest energies. At each longitudinal position, ten images were taken and the FWHM beam size derived from the images are plotted in Figure 53. At 75 μJ (Figure 53a), a single filament blocked.
was clearly observed. Self-cleaning of the beam profile occurred between $z = 1.85$ m and 1.9 m, and the profile exhibited a relatively stable beam size (between 80 μm and 100 μm) and a high level of symmetry between $z = 1.9$ m and 2 m.

When the energy was increased to $173 \mu$J (Figure 53b), the beam shrunk more rapidly horizontally, but some energy remained dispersed vertically. The horizontal beam size remained stable (between 90 μm and 110 μm) from $z = 1.87$ m to close to 2 m. The beam acquired a circularly symmetric core, but the energy surrounding it was highly irregular in shape. Past the geometric focus, the beam broke up and the profile became irregular. This behavior was characteristic of the filament at energies between 150 μJ and 200 μJ. This is the highest energy level at which we would consider the beam to be a single filament.

When the energy was increased to $300 \mu$J (Figure 53c), multi-filament-like structures could be observed even for $z < 1.8$ m. The energy distribution evolved rapidly with propagation, and the profile broke up past the geometric focus. The size of each filament core that developed was $\lesssim 100$ μm.
Figure 54. Comparison between the peak fluence recorded by the grazing incidence imaging system and the level of plasma emission in the images in Figure 50.

From the profiles taken using the grazing incidence imaging system, it was possible to track the evolution of the peak fluence in the beam with propagation distance. In Figure 54, the peak fluence is compared to the plasma emission from the images in Figure 50. It can be observed that the fluence levels and the plasma emissions both reach their maxima at approximately the same location for each pulse energy. This demonstrates good consistency between the information gathered based on the beam profiles and that from the images of the plasma emission.
4.3.2.3 - Spectral Broadening

Figure 55. Spectrum of the UV pulse after filamentation with 2 m focusing. The cross sections in the colormap (a) defined by the dashed lines are plotted together in (b).

The same setup used to measure the spectrum of the UV pulse in Figure 49 was used to measure the spectrum of the pulse after filamentation. The ground CaF$_2$ plate was placed 2.7 m from the lens where filamentation has ceased and the plate was not damaged by the beam. Spectral broadening began once the peak power exceeded $P_{cr}$ (10 μJ pulse energy). The spectrum broadened by a similar amount on both sides of the original spectrum, although the spectral peak shifted towards longer wavelengths with increasing energy.
Figure 56. Spectrum of the UV pulse after filamentation with 10 m focusing. The cross sections in the color maps (a,c) defined by the dashed lines are plotted together in (b,d). The same information is represented in (a,b) in logarithmic scale, and (c,d) in linear scale.

At 800 nm, a longer geometric focal distance leads to longer filaments and consequently greater spectral broadening. To verify if this is true for UV filaments as well, the measurement of the post-filamentation spectrum was repeated with a lens of 10 m focal distance, with the diffusing plate at 10.75 m. The results in Figure 56 show that the spectrum was indeed broader with a longer focal distance, and also revealed several interesting features that were not present for the shorter focal distance. Figure 56a,b again shows that the amount of broadening was again fairly similar on both sides, and the spectra
were similarly biased towards the longer wavelengths. However, the spectrum appeared to stop extending to the shorter wavelengths for energies above 200 µJ and cut off below 256 nm, while broadening to the longer wavelengths was not affected.

For the high intensity structures in the spectrum to be more visible, the data was replotted in the linear scale (Figure 56c,d). The spectrum developed sharp intensity peaks, or spectral fringes, that shifted towards longer wavelengths with increasing energy. The first peak could be seen to appear between 26 µJ and 53 µJ. This peak continued to shift towards 275 nm with higher energy, and new peaks appeared at 159 µJ, 337 µJ and 468 µJ near 267 nm. As the data is not sufficiently dense to create a colormap that accurately represents the movement of the peaks, dotted green lines have been added in Figure 56c to guide the eye. The peaks were regularly spaced, with an average of 1.95 nm between them. Tzortzakis et al. have observed similar spectral peaks/fringes in the post-filament spectrum [184] of 450 fs pulses at 248 nm, which they deduced to be the beat frequency between the split pulses in the filament. In our case, a 1.95 nm interval between peaks imply a beat frequency of 8.2 THz, or split pulses 122 fs apart in time.

Figure 57. Fluorescence generated on a sheet of paper by the post-filament beam at 10.75 m distance. The scale is indicated by the millimeter markings below the beam.
A further observation was made in the 10 m focusing condition. With a sheet of printer paper placed in the beam, the fluorescence of the paper when excited by UV radiation can reveal the transverse extent of the beam. Figure 57 shows the fluorescence generated by the beam at different pulse energies. More UV energy was dispersed radially outwards as the energy was increased, far beyond the geometrical beam size if the beam had been propagating linearly (leftmost image at 6.9 μJ). This could be evidence of conical emission in the UV generated by the filament, or low intensity energy at the end of the pulse that had been defocused and diffracted by the plasma, and that did not have sufficient power to refocus and participate in the filamentation process. The speckled appearance of the fluorescence, even at 53 μJ where only a single filament should have formed, suggests strong interference taking place. This is different from single 800 nm filaments, which typically have a highly symmetric appearance post-filamentation. A possible explanation is the distortion of the wavefront by the low density plasma pedestal surrounding the filament core described by Fedorov and Kandidov [188], which in reality would not have a smooth and circularly symmetric profile.

4.3.3 - Simulation of UV Filaments

The filamentation simulation code described in Chapter 2.2.1 was used to numerically determine the outcome of filamentation of a UV pulse. Certain parameters were changed from those discussed in Chapter 1.1 to account for the properties of air at 267 nm. The nonlinear refractive index and dispersion were $n_2 = 8 \times 10^{-19}$ cm$^2$/W and $k'' = 1.21$ fs$^2$/m. Oxygen was verified to still be the dominant ionizing species, and the 3-photon...
ionization cross section was $\sigma_3 = 1.91 \times 10^{-28}$. To achieve the spatial resolution required for UV filaments without significantly increasing the simulation time, the initial beam size used was 2 mm FWHM (instead of 4-5 mm as measured). To maintain a similar NA, the focal distance simulated was 1 m. The initial positive chirp of the pulse was also taken into consideration, and the actual pulse duration used was 75 fs, with a theoretical transform limited duration of 50 fs.

4.3.3.1 - Simulation Results with $4.7P_{cr}$ and 1 m focus

Figure 58. Simulation results of UV filament with 50 μJ energy and 1 m focusing distance. The first simulation was performed with a 50 μJ pulse, and the results are shown in Figure 58. The filament was 8 cm in length, based on the zone where the fluence FHWM of the beam showed that the beam ceased to converge and before beam divergence began. The simulated FWHM in the filament ranged between 45-60 μm. The simulated filament length
compares well with that observed in the laboratory (Figure 51), but the simulated filament diameter was around 50% smaller than the measured values (Figure 53). Based on the numerical results, at $f = 2$ m UV filaments would be noticeably smaller than 800 nm filaments, even though the measurements suggest that the sizes should be similar (see Figure 12).

The peak intensities reached were similar between the UV and 800 nm filaments, while the plasma channel in the UV filament was ~10 times as dense as that of the 800 nm filament.

The temporal profile showed pulse splitting occurring; however, the pulse splitting behavior is qualitatively different from that of filaments in the NIR. At 800 nm, the pulse initially appears to move forward in time in the group velocity reference frame, and after propagating for a certain distance, a second distinct pulse appears behind the first due to energy at the rear of the pulse (which has been defocused by the plasma) refocusing back on-axis (see e.g. Figure 13, Figure 15 and Figure 32). For the UV filament, the second pulse appear to split directly and immediately from the first pulse. Such behavior is characteristic of pulse splitting due to ionization losses rather than defocusing and refocusing effects [163, 184]. The high intensity peak of the pulse is strongly absorbed in the photoionization process, causing the front and rear of the pulse to appear as two split pulses. In the results in Figure 58, the weaker second pulse can be explained by the defocusing and absorption by the plasma generated, which the first pulse is not subjected to.
To obtain comparisons similar to the measurement results presented in Chapter 4.3.2, two more simulations were performed – one with 150 μJ energy and 1 m focusing distance (Figure 59), and the other with 50 μJ energy and 5 m focusing distance (Figure 60).

**4.3.3.2 - Simulation Results with 14P_{cr} and 1 m focus**

Figure 59. Simulation results of UV filament with 150 μJ energy and 1 m focusing distance.

For the simulation with higher energy, the filament length maintained good agreement with the data in Figure 51. The filament size at the beginning was ~70 μm, and gradually decreased to ~45 μm at the geometrical focus. The intensity and plasma density were slightly higher than the 50 μJ case. The temporal profile exhibited complex pulse splitting behavior, with the leading pulse persisting for the full length of the filament while trailing pulses evolved rapidly.
4.3.3.3 - Simulation Results with $4.7P_{cr}$ and 5 m focus

For the simulation with 5 m focusing distance, refocusing of the beam ~0.7 m from the position where filamentation first begun was observed, forming two intensity peaks. The first collapse zone reached a minimum beam size of ~52 μm while the second was ~76 μm. In between the two collapse zones, the beam stayed smaller than 115 μm. The highest plasma density obtained was an order of magnitude lower than in the 1 m focusing case. The temporal profile in this case showed near-symmetric pulse splitting, indicating that MPI loss is clearly a dominant physical process [163, 184]. It is responsible for creating the split pulses, and possibly arresting the collapse of the self-focusing beam as well. The first pair of split pulses reached a separation of ~125 fs at the position $z = 4.75$ m, which matches well with the deduction from the measured spectral peak interval (see Chapter 4.3.2.3).
4.3.3.4 - Spectral Features from Simulation

Figure 61. Comparison between (a,b,c) simulated and (d,e,f) measured spectra. The original spectrum of the pulse is given by the dash-dot black line. The simulated spectra were taken only from the on-axis portion of the beams.

The spectra from the simulations were compared to those measured in the laboratory. For easier comparison, the focusing conditions are indicated in NA in Figure 61. To obtain spectra with comparable features, the experimental spectra were taken from more energetic pulses. Many of the features in the measured spectra were reproduced in the simulations: the strong single peak in Figure 61a and d, the split spectral peak in Figure 61b and d, and the ~2 nm spacing between maxima in Figure 61c and f. However, the spectra from simulation did not exhibit a significant redshift like the experimental spectra. The simulated spectrum in the low NA case (Figure 61c) had broad peaks on both sides of
the spectrum, which is characteristic of broadening by SPM. These peaks were not present in the measured spectrum.

![Figure 62](image)

Figure 62. Comparison between the on-axis (blue solid lines) and whole beam integrated (red dashed lines) spectra.

It should be noted that these features in the simulated spectra are only visible on-axis. If the spectra were integrated across the whole beam, the results were a lot more featureless (Figure 62). This is because the low intensity periphery of the beam and the conical emission do not undergo the same temporal evolution as the filament core. This also confirms that the spectral measurements were made primarily on the center of the beam, and the periphery of the beam was not collected by the fiber.

4.3.4 - Discussion

The grazing angle imaging system provided a simple and straightforward means of observing the spatial evolution and the size of the UV filaments. In these observations, the FWHM diameter of the filaments were consistently ~100 μm for different pulse energies. However, numerical results produced filaments that were ~25-50% smaller. Nonetheless,
such differences between experimentally and numerically determined filament sizes are not uncommon. Table 3 shows measured UV filament sizes to be between 100-200 μm, while simulated sizes in the literature [18, 185, 187] are typically <100 μm. The values obtained in this study are therefore within the typical ranges for UV filaments.

The variations in the spectral characteristics of UV filaments are greater with different pulse durations. Schwarz et al. [29] observed effectively no spectral broadening with picosecond UV pulses, while Tzortzakis et al. [184] observed broadening only towards longer wavelengths with 450 fs pulses. With 75 fs pulses, we have observed broadening on both sides of the spectrum, but with a redshift of the spectral energy. This redshift, though present in the measurements of Tzortzakis et al., has not been previously discussed. For filaments at 800 nm, Chen et al. have shown that a second red-shifted spectral peak appears for filaments with sufficient energy and length [193]. The second peak appears over the course of filamentation, and shifts further towards longer wavelengths with propagation. The red-shift is faster with higher energy, and stops once the filament terminates. Uryupina et al. demonstrated that the red-shifted peak is the result of the Raman shifted component forming a soliton in the filament, supported by the negative GVD in a sufficiently dense plasma channel [194]. The red-shift observed in the UV filaments could therefore be from the same effect. The relative magnitude of the red-shift away from the central wavelength could be different in the UV due to the different dispersion characteristics of air at 267 nm and at 800 nm, and the different characteristics of the filaments formed.
Considering that the UV supercontinuum exhibits spectral modulation which accurately reflects the pulse-splitting phenomenon, one may wonder if a similar observation could be made for NIR filaments. For a similar delay between the split pulses, the spectral fringe spacing around 800 nm could be deduced to be 9x greater, i.e. around 20 nm. The spectral content would be relatively weak at such distances from the central wavelength (820 nm, 840 nm, etc.), making the observation of these fringes difficult. The Raman red-shifted peak described by Chen et al. and Uryupin et al. [193, 194] may also mask the appearance of such spectral modulation in the NIR.

MPI is clearly a dominant physical process in UV filamentation. For focused beams with higher NA, it works in tandem with plasma defocusing in limiting the increase in intensity and the collapse of the beam, while for lower NA cases MPI becomes the main intensity limiting mechanism. This is deduced from the pulse splitting behavior seen in the simulation results, and confirmed by the appearance of spectral fringes in the measured spectra, whose spacing matches well with the beat frequency of the split pulses. This was expected based on the results in Chapter 3.2, where MPI loss was deduced to play a dominant role in the 3-photon ionization regime. Due to greater MPI losses, the length of each continuous zone of high intensity and plasma density in a UV filament is therefore shorter. This is qualitatively observable in the simulation results with low-NA focusing (compare Figure 15 for the 800 nm case and Figure 60 for the 267 nm case), as well as in the results presented by Couairon and Bergé [18]. Conversely, the plasma density does not
diminish as drastically between refocusing events in UV filaments. This could be important in applications where high plasma densities is not as important as maintaining the continuity of the plasma channel. The possibility of increasing the plasma density to obtain a continuous high density channel via an igniter-heater scheme [55, 56] could also be considered in this case. A consequence of MPI being a dominant process in UV filamentation is that the focusing regimes described in Chapter 4.1 are no longer valid. This was already evident in the three simulation results presented in this chapter, where significant differences existed in the intensity and plasma density values even though they should have all belonged to the nonlinear-focusing regime.

4.3.5 - Summary

UV filaments from sub-100 fs pulses generated by THG from a Ti:sapphire laser have been examined. The beam profiles of the UV filaments and the supercontinuum generated have been carefully measured. Together with the simulation results, they lead us to the conclusion that filamentation in the UV (in the 3-photon ionization regime) is based on a different balance of physical processes from filamentation at 800 nm. MPI, instead of plasma defocusing, is the key mechanism arresting the collapse of the beam. Tzortzakis et al. had reached the same conclusion for UV filamentation with picosecond and sub-picosecond pulses [184], indicating that the physics governing filamentation in the UV remains unchanged despite two orders of magnitude difference in the pulse duration.
CHAPTER 5 – CONCLUSION

The impact of several fundamental conditions and parameters on the filamentation process has been carefully investigated, and the results have led to a more profound understanding of filamentation science in general. The two tools developed as part of this dissertation, the grazing incidence imaging system and the NLSE filament simulation code, have proven to be versatile and effective. The imaging system has allowed the fluence profile of a filament to be measured with a high level of confidence, and the simulation code provided results that complemented and gave stronger credence to the experimental findings.

A large majority of the applications of filamentation require the propagation of the filament over long distances through the atmosphere. For the findings in this work to be most directly relevant and applicable, it would have been desirable for all the investigations to be performed on filaments propagating in air. However, limitations in the type of sources currently available required that certain experiments be conducted in a different medium. In the first experiment on filamentation in solids, the effective three-wave mixing model was used to explain the generation of a strong blueshifted spectral peak in the anomalous dispersion regime. With the continued development of high power sources in the mid-IR, similar observations may one day be made in air. The second experiment demonstrated that the fewer photons are required for multi-photon ionization, the more multi-photon absorption will have an impact on filamentation. This conclusion was subsequently validated in air using pulses in the UV.
The experiments in air and other gases culminated in separate conclusions on the effects of focusing condition, polarization, and switching from NIR to UV pulses. The first experiment dealt with geometrical focusing conditions. Using a simplified analytical model that we derived, filamentation can be classified as occurring in the linear-focusing or nonlinear-focusing regime. The transition point between the two regimes was clearly defined. The balance between the physical processes is altered when moving from one regime to the other, making the transition highly relevant to laboratory-based experiments, which typically involve focused beams. The second experiment examined polarization effects. By changing the initial polarization of the beam between linear and circular polarizations, an anomalous spectral broadening behavior was observed for specific elliptical polarizations. Further investigation allowed us to determine that the anomalous behavior originated from the molecular dynamics of the gaseous medium. This finding could lead to better control and enhancement of the supercontinuum generation process, potentially benefiting applications that exploit this property of laser filaments. The third experiment looked at filaments generated by the frequency-tripled beam from the 800 nm source. Filamentation of UV pulses obeys a different hierarchy of participating physical mechanisms – MPI becomes the primary intensity limiting mechanism that prevents the collapse of the beam. Even with such a fundamental change to the physics when switching between UV and NIR wavelengths, the variations in many of the filament’s physical properties are relatively small.
The results presented in this dissertation have demonstrated the complexity of filamentation science. The basic principle of filamentation is well understood: the interplay between self-focusing and a counteracting effect (plasma defocusing or MPI) allows the beam to maintain a relatively constant profile as it propagates. However, there are many parameters involved in the process that still warrant closer examination. Some, such as the focusing condition, just required better characterization and definition. Others, such as polarization, have effects that we can only partially explain within the framework of this dissertation. Nonetheless, the findings in this dissertation will serve as a step towards the community’s eventual complete mastery of this fascinating physical phenomenon that is filamentation, and hopefully its deployment in real-world applications.
The Multi-Terawatt Filamentation Laser or Multi-Terawatt Femtosecond Laser (MTFL) system is a Ti:sapphire chirped pulse amplification (CPA) system capable of delivering ultrashort pulses with sufficient energy for long range, multi-filament applications.

The MTFL system consists of many sub-systems, see diagram and descriptions below.

This appendix is an excerpt of the MTFL Operation Manual. More details about the operation and maintenance of the system can be found there.

Figure 63. Schematic layout of the MTFL system on a 16' x 4' optical table.
MTFL outputs

Three different outputs are possible:

- **Kilohertz line**: 1 kHz repetition rate, 50 fs pulse duration, 1 mJ pulse energy
- **1” line**: 10 Hz repetition rate, <45 fs pulse duration, 10 mJ pulse energy
- **Full-energy line**: 10 Hz repetition rate, <45 fs pulse duration, 470 mJ pulse energy

The 1” line is given its name because it allows the use of 1” optics (including 45° mirrors).

MTLF sub-systems

Oscillator

The oscillator is a Spectra-Physics Tsunami mode-locked Ti:sapphire laser pumped by a Spectra-Physics Millenia V CW 532 nm laser. The oscillator puts out an 74 MHz pulse train of 35 fs, 800 nm pulses, with an average power of around 330 mW.

Pulses from the oscillator are picked at 1 kHz (or 10 Hz) to seed the regenerative amplifier. The emitted bandwidth from the oscillator must be kept sufficiently broad to ensure an optimal final compressed pulse at the MTFL output, and the emitted average power must be sufficiently high (<300 mW) to ensure stable operation of the regenerative amplifier.
Both the Millenia V and Tsunami are cooled by a Lytron recirculating chiller (the smaller one in the chaseway) set to 17°C.

For more information on the Millenia V and the Tsunami, refer to the respective binders.

Stretcher

The stretcher is an in-house designed Offner type pulse stretcher with a single 1800 lp/mm grating. Using 2 passes (4 reflections per pass) off the grating, the oscillator pulses are stretched to ~450 ps in duration.
The stretcher grating has 1400 lp/mm, and the convex and concave mirrors have 500 mm and 1000 mm curvatures respectively.

A de-magnifying telescope at the output of the stretcher reduces the beam size for transmission through the Dazzler, isolator and for seeding the regenerative amplifier.

**Dazzler**

The Dazzler is an Acousto-Optic Programmable Dispersive Filter developed by Fastlite that provides control over the spectral phase and amplitude of the seed pulse entering the regenerative amplifier. To counter spectral gain narrowing effects in the amplifier stages, the Dazzler is used to insert a dip in the seed spectrum near the center (800 nm). To
correct for dispersion effects in the system, a file containing spectral phase information is loaded into the Dazzler during operation.

![Image of beam path and Dazzler components]

Figure 66. Beam path between stretcher output and the regen. The wedge allows the spectrum after the Dazzler to be inspected if necessary. Source (right): http://www.fastlite.com/en/cat465015--Dazzler.html

Depending on the desired output, different Dazzler presets need to be loaded for optimal pulse duration and spectral content. The Dazzler is controlled by the main control computer. The Dazzler preset files are saved in the folder “V420Q-Dir”.

For more information on the Dazzler, refer to the manual provided by Fastlite.

**Regenerative Amplifier**

The regenerative amplifier (or “regen”) is a Spectra-Physics Spitfire amplifier system (minus the stock stretcher and compressor). The regen gain medium is a Ti:sapphire
crystal double-side pumped by a Spectra-Physics Evolution-30 pulsed laser emitting 20 W of 527 nm pulses at 1 kHz. The regen cavity is Z-shaped, with two Pockels cells to time the entry of the seed pulse into the cavity and to eject the amplified pulse. In ~15 to 17 round trips (each round trip ~10 ns) in the cavity, the seed pulse energy is amplified to ~2.5 to 3 mJ. The ejected beam is sent through a telescope (f = -38 mm and f = +150 mm) to increase the beam size.

Figure 67. Regenerative amplifier. Solid pink line marks the Z-shaped cavity path, dashed pink line the seed path, dotted pink line the regen output, and green line the pump.

The regen crystal is TEC chilled to 0°C. To prevent condensation from forming on the crystal when it is being cooled, the crystal box needs to be purged with dry nitrogen when the regen is operating. The Evolution-30 and the three amplifier (regen, pre-amp, final amp) crystal mounts are cooled by a Lytron recirculating chiller (the larger one in the chaseway) set to 21°C.
A fixed iris has been introduced into the regen cavity to help improve the beam quality. A thin film polarizer after the output of the regen helps to reject leaked pre-pulses from the cavity. A light valve after that allows the output energy to be controlled. The regen can be operated at 1 kHz or at 10 Hz.

To ensure that the subsequent amplifier stages are seeded with the best beam profile possible, the output of the regen goes through a spatial filter consisting of a +400 mm focusing lens, a 0.1 mm pinhole and a +250 mm re-collimating lens. The beam size and divergence is also modified at this stage for optimal operation of the pre-amplifier.

A flip mirror can be raised to direct the beam towards a telescope (to resize the beam and remove the intentional beam divergence) and then to the kHz compressor.

For more information on the regen, refer to the relevant Spitfire binders provided by Spectra-Physics.

kHz Compressor

The kHz compressor is a Treacy compressor with two 1500 lp/mm gratings. Grating 2 is mounted on a motorized translation stage with 1” of travel.
The kHz compressor is built on a removable breadboard to facilitate access to the rest of the system, and to allow the table space to be liberated when necessary. The breadboard sits on a set of kinematic mounts (Thorlabs KBS98) that allows accurate and repeatable repositioning. Nonetheless, the quality of the compressed output (spatial chirp, grating position for best compression, spectral phase, etc.) should be verified each time the kHz compressor is removed and replaced.

Pre-amplifier

The pre-amp is a 6-pass bowtie amplifier, with an uncoated, normal incidence Ti:sapphire crystal 10 mm in diameter and 15 mm in length. The crystal is pumped on both sides by the output of the Spectra-Physics Quanta-ray Pro 290. The Quanta-ray emits ~750 mJ pulses at 532 nm. A light valve mounted at the output of the Quanta-ray controls the amount of pump energy incident on the pre-amp crystal. The pump beam is split by a beamsplitter, and the SHG crystal of the Quanta-ray is relay imaged onto the Ti:sapphire crystal by both beams from opposite sides. Vacuum relay tubes with Brewster windows ensure that there is no loss of energy and distortion of the relay images by air-breakdown plasmas.
The seed beam for the pre-amp is slightly divergent. This is a way of combatting the spatial gain narrowing and thermal lensing effects in the amplification process. Even with the current diverging seed beam, the output of the pre-amp is slightly convergent. At the end of six passes, the seed beam is amplified from \(\sim 2.5 \text{ mJ} \) to \(\sim 160 \text{ mJ} \).

For more information on the Quanta-ray, refer to the binder provided by Spectra-Physics.

**Spatial Filter**

The output of the pre-amp is sent into a spatial filter to clean up its beam profile for the 1" line output or for further amplification in the final amp. The spatial filter consists of a 500 mm focusing lens, a 0.2 mm pinhole and a 700 mm re-collimating lens. Most of the propagation between the two lenses occurs in a vacuum tube (\(10^{-6} \text{ torr maintained by an ion pump}\)) to prevent the breakdown of air at the pinhole.
The input to the spatial filter is slightly convergent; the lenses of the spatial filter have been set to re-collimate the beam. The spatial filter has a total transmission of \( \sim 70\% \), therefore the output energy is \( \sim 112 \text{ mJ} \).

A motorized flip mirror allows the output of the spatial filter to be directed towards the final amp (flip mirror down) or the high-energy compressor for the 1” line output (flip mirror up). When the flip mirror is up, the beam is first sent into a light valve and attenuator (see Figure 71) then a telescope (-200 mm and +300 mm lenses) to increase the beam size for compression. The enlarged beam is then directed into the high-energy compressor by a periscope and a flip mirror.
Figure 71. Light valve and attenuator setup. The half-wave plate and polarizing beamsplitter function as a light valve, while the fused silica wedge in the middle caps the transmission of the setup at 18%. This prevents the 1” line beam from reaching high fluences which will damage the compressor grating.

Final Amplifier

The final amp is a 3-pass bowtie amplifier with an uncoated, normal incidence Ti:sapphire crystal 16 mm in diameter and 20 mm in length. The crystal is pumped on both sides by the output of the Continuum Powerlite DLS. The Powerlite emits ~2 J pulses at 532 nm. The overall considerations for pumping and seeding the final amp are similar to those of the pre-amp. Before the seed enters the final amp, it is expanded by a telescope (-250 mm and +400 mm) to provide the necessary beam size and divergence. At the end of three passes, the seed beam is amplified from ~112 mJ to ~750 mJ.
Figure 72. Pictures of the final amp bowtie. The mirrors M1 to M10 are labeled in order, from the 1\textsuperscript{st} pass to the 3\textsuperscript{rd} pass. The pink lines indicate the input and output beam paths. The center picture shows the Ti:sapphire crystal in its mount.

After the third pass, the beam is elevated by a periscope and expanded by another telescope (-200 mm and +600 mm) before it is directed into the high-energy compressor.

The beam size after this telescope is large and requires the use of 2” optics (at normal incidence) and 3” mirrors.

For more information on the Powerlite, refer to the manual provided by Continuum.

High-energy Compressor

The high-energy compressor is a Treacy compressor with two 1800 lp/mm gratings. Grating 2 is mounted on a motorized translation stage with 2” of travel.
The high-energy compressor is built in a vacuum chamber, allowing the compression to be carried out in a vacuum down to $10^{-5}$-$10^{-6}$ torr. There were initial concerns about build up of carbon deposits due to the high fluences in the compressor. It was assumed that a vacuum environment would minimize this build up. However, that was concluded not to be the case (think other filamentation lasers eg. Teramobile, ENSTAmobile, T&T, etc. and also communication with Lund University).
Pulse compression for filamentation experiments is therefore carried out in atmosphere, but the compressor may still be pumped down for other types of experiments. A turbo pump is connected to the compressor chamber and the chamber can be pumped down to ~$10^{-5}$ torr if required.

**Triggering and Timing Devices**

The entire system needs to be accurately timed and synchronized for the correct amplification to be achieved. Suboptimal synchronization can result in poor amplification, over-amplification (potentially damaging certain components) and/or poor stability.

The master clock for the system is a homemade frequency divider circuit. It receives the 75 MHz pulse train from the Tsunami, and divides that to a synchronized 1 kHz output and
a switchable 1 kHz or 10 Hz output. These signals are then sent as trigger signals to various delay generators to create the necessary triggers for the different sub-systems of the MTFL.

Three different Stanford Research Systems DG535 delay generators control most of the trigger signals in the system. The optimal settings for the DG535s are stored in memory “8”. The Spectra-Physics SDG II is a high-voltage supply and delay generator for the regen Pockels cells. For more information on the SDG II, refer to the binder provided by Spectra-Physics. Information on the delay settings can be found in the Operation Manual.

Figure 75. Timing and synchronization scheme for MTFL.

**Cooling System and Plumbing**

There are two Lytron recirculating chillers and a ThermoNESLAB water/water heat exchanger cooling the MTFL system, located in the chaseway behind the laboratory. The key to the chaseway is kept in the third drawer of the black filing cabinet behind the MTFL table, in a key storage box. The #8 tag is attached to the chaseway key.
The small Lytron chiller cools the Millennia V and the Tsunami (connected in series). There is a by-pass built into the source and return lines to lower the pressure in the lines. This chiller is set to 17°C.

The larger Lytron chiller cools the Evolution 30 and the regen, pre-amp and final amp Ti:sapphire crystals (connected in series). A filter is attached to the source line to keep the water flowing through the system clean. This chiller is set to 21°C.

The ThermoNESLAB heat exchanger removes heat from the built-in reservoirs of the Quanta-ray and the Powerlite (connected in parallel) and dumps it on the university chilled water lines. The thick orange hoses lead to the Quanta-ray and the thinner transparent
hoses lead to the Powerlite. There is a pressure regulator connecting the source and return lines to keep the pressure at 60 psi.
APPENDIX B - SIMULATION PARAMETERS
This appendix is a compilation of the parameters used for the simulation of filamentation as described in Chapter 2.2. As some of the filamentation simulation done are out of the scope of this dissertation and not all simulation results have not been presented in this document, some of the parameters listed in this appendix may not seem to be directly pertinent. Where possible, the sources of the parameters are cited.

Key Variables and Equations

Electric Field

\[ E(\mathbf{r}, t) = \sqrt{\frac{\omega_0 \mu_0}{2k_0}} \varepsilon(\mathbf{r}, t)e^{i(k_0 z^2 - \omega_0 t)} + c.c. \] \hspace{1cm} (36)

Initial Pulse (assumed cylindrically symmetric)

\[ \varepsilon(r, t, z = 0) = \varepsilon_0 e^{-\frac{r^2}{\sigma_{\tau}^2}} e^{-i\frac{k_0 z^2}{2F^2}} e^{-\frac{t^2}{\sigma_{\tau0}^2}} \] \hspace{1cm} (37)

\[ \varepsilon_0 = \sqrt{\frac{2}{\pi} \frac{P_0}{\sigma_{\tau0}^2}} \] \hspace{1cm} (38)

\[ P_0 = \sqrt{\frac{4 \ln 2 \text{Eng}}{\pi \tau}} \] \hspace{1cm} (39)

Here, \( F \) is the initial geometrical focal distance, \( \text{Eng} \) is the pulse energy, and \( \tau \) is the intensity FWHM pulse duration.

Chirped Pulse

\[ \varepsilon(r, t, z = 0) = \varepsilon_0 e^{-\frac{r^2}{\sigma_{\tau}^2}} e^{-i\frac{k_0 z^2}{2F^2}} e^{-(1+ic)\frac{t^2}{\sigma_{\tau c}^2}} \] \hspace{1cm} (40)
Here, \( \sigma_{t,c} \) is the chirped pulse duration while \( \sigma_{t0} \) is the transform limited pulse duration.

The NLSE (Equation (9)) and the ionization rate equation (Equation (6)) have been described in Chapter 1.1.

**Parameters for 800 nm**

Table 4. List of parameters used for filamentation simulation at 800 nm.

<table>
<thead>
<tr>
<th></th>
<th>Air</th>
<th>( N_2 )</th>
<th>( O_2 )</th>
<th>( CO_2 )</th>
<th>( Ar ) [j]</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>( n_2 ) ( \times 10^{-19} ) cm(^2)/W</td>
<td>2.9</td>
<td>2.3</td>
<td>5.1</td>
<td>3</td>
<td>3</td>
<td>[a]</td>
</tr>
<tr>
<td>( k^* ) [fs(^2)/cm]</td>
<td>0.2005</td>
<td>0.187</td>
<td>0.2476</td>
<td>0.309</td>
<td>0.1798</td>
<td>[b]</td>
</tr>
<tr>
<td>( \sigma_K ) [s(^{-1})cm(^2)KW(^{-4})]</td>
<td>See ( O_2 )</td>
<td>6.31</td>
<td>2.81</td>
<td>( 5 \times 10^{-140} )</td>
<td>[c]</td>
<td></td>
</tr>
<tr>
<td>( \times 10^{-140} ) ( \times 10^{-96} )</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( U_i ) [eV]</td>
<td>See ( O_2 )</td>
<td>15.576</td>
<td>12.063</td>
<td>13.7 [d]</td>
<td>15.76</td>
<td>[c]</td>
</tr>
<tr>
<td>( K ) (1.55 eV @ 800 nm)</td>
<td>See ( O_2 )</td>
<td>11</td>
<td>8</td>
<td>9</td>
<td>11</td>
<td>[c,e]</td>
</tr>
<tr>
<td>( \Gamma^{-1} ) [fs]</td>
<td>70</td>
<td>70</td>
<td>70</td>
<td>180 [f]</td>
<td>N.A.</td>
<td>[c,e]</td>
</tr>
<tr>
<td>( B_e ) [cm(^{-1})]</td>
<td>2.01</td>
<td>1.438</td>
<td>0.3915</td>
<td>N.A.</td>
<td>[g]</td>
<td></td>
</tr>
<tr>
<td>( J )</td>
<td>7 or 9</td>
<td>8</td>
<td>16</td>
<td>N.A.</td>
<td>[h]</td>
<td></td>
</tr>
<tr>
<td>( \omega_r ) ( \times 10^{12} ) s(^{-1})]</td>
<td>16 [c]</td>
<td>14.4</td>
<td>10.3</td>
<td>5.16</td>
<td>N.A.</td>
<td>[i]</td>
</tr>
</tbody>
</table>


[d] [http://webbook.nist.gov/cgi/cbook.cgi?ID=C124389&Mask=20](http://webbook.nist.gov/cgi/cbook.cgi?ID=C124389&Mask=20)


[f] Private communication – simulations by Ramakrishna Sesha Shankar (Northwestern University)
population = \((2J + 1)e^{-\frac{\hbar c B_e J(j+1)}{k_B T}}\), find most populated \(J\) state

\[\omega_r = 4\pi B_e c (2J + 3), \text{ Ripoche et al., Opt. Commun. 135, 310 (1997)}\]


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### Parameters for UV Filaments in Air

Table 5. List of parameters in the literature for UV filaments in air.

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>(n_2 \times 10^{-19} \text{ cm}^2/\text{W})</td>
<td>8</td>
<td>8</td>
<td>13.4 (they're high for 800nm as well)</td>
</tr>
<tr>
<td>(k'' \text{ [fs}^2/\text{cm]})</td>
<td>No dispersion</td>
<td>1.2</td>
<td></td>
</tr>
<tr>
<td>Oxygen</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(\sigma_K / s^{-1} \text{ cm}^2 \text{ W}^{-K} )</td>
<td>12.06</td>
<td>PPT, has rate plot</td>
<td></td>
</tr>
<tr>
<td>(K (4.65 \text{ eV} @ 266 \text{ nm}))</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Nitrogen</td>
<td>3</td>
<td>PPT, has rate plot</td>
<td></td>
</tr>
<tr>
<td>(\sigma_K / s^{-1} \text{ cm}^2 \text{ W}^{-K} )</td>
<td>15.58</td>
<td>Ignored, with ref.</td>
<td></td>
</tr>
<tr>
<td>(K (4.65 \text{ eV} @ 266 \text{ nm}))</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Raman</td>
<td>4</td>
<td>PPT, has rate plot</td>
<td></td>
</tr>
<tr>
<td>(\Gamma^{-1} \text{ [fs]})</td>
<td>Not included, no red-shift,</td>
<td>70 fs</td>
<td>Ignored</td>
</tr>
<tr>
<td>(B_e \text{ [cm}^{-1}])</td>
<td>indicate Raman-Kerr small.</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(\omega_r \times 10^{12} \text{ s}^{-1})</td>
<td>70 fs</td>
<td></td>
<td></td>
</tr>
<tr>
<td>--------------------------</td>
<td>--------------------------</td>
<td>--------------------</td>
<td>-----------------------------</td>
</tr>
<tr>
<td>$n_2 [\times 10^{-19} \text{ cm}^2/\text{W}]$</td>
<td>10</td>
<td>7.8</td>
<td>13.4 (they're high for 800nm as well) Sellmeier</td>
</tr>
<tr>
<td>$k'' \text{ [fs}^2/\text{cm]}$</td>
<td>2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Oxygen</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\sigma_K [s^{-1} \text{cm}^2 K \text{W}^{-K}]$</td>
<td>$2.5 \times 10^{-28}$</td>
<td>$3 \times 10^{-35}$</td>
<td>$1.34 \times 10^{-27}$</td>
</tr>
<tr>
<td>$U_i [\text{eV}]$</td>
<td></td>
<td>12.2</td>
<td></td>
</tr>
<tr>
<td>$K (4.65 \text{ eV} @ 266 \text{ nm})$</td>
<td>3</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>Nitrogen</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\sigma_K [s^{-1} \text{cm}^2 K \text{W}^{-K}]$</td>
<td>Ignored</td>
<td>Ignored</td>
<td>$2.4 \times 10^{-43}$</td>
</tr>
<tr>
<td>$U_i [\text{eV}]$</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$K (4.65 \text{ eV} @ 266 \text{ nm})$</td>
<td>Ignored</td>
<td>Ignored</td>
<td></td>
</tr>
<tr>
<td>Raman</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\Gamma^{-1} \text{ [fs]}$</td>
<td>26 THz</td>
<td>Ignored</td>
<td>52 THz (?)</td>
</tr>
<tr>
<td>$B_e \text{ [cm}^{-1}]$</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$f$</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\omega_r \text{ [}\times 10^{12} \text{ s}^{-1}]$</td>
<td>16 THz</td>
<td>16 THz</td>
<td></td>
</tr>
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</table>

<table>
<thead>
<tr>
<th></th>
<th>Couairon, PRL 2002</th>
</tr>
</thead>
<tbody>
<tr>
<td>$n_2 [\times 10^{-19} \text{ cm}^2/\text{W}]$</td>
<td>8</td>
</tr>
<tr>
<td>$k'' \text{ [fs}^2/\text{cm]}$</td>
<td>1.21</td>
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<td>Oxygen</td>
<td></td>
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<td>$\sigma_K [s^{-1} \text{cm}^2 K \text{W}^{-K}]$</td>
<td>$1.91 \times 10^{-28}$</td>
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<td>$U_i [\text{eV}]$</td>
<td>12.1</td>
</tr>
<tr>
<td>$K (4.65 \text{ eV} @ 266 \text{ nm})$</td>
<td>3</td>
</tr>
<tr>
<td>Nitrogen</td>
<td></td>
</tr>
<tr>
<td>$\sigma_K [s^{-1} \text{cm}^2 K \text{W}^{-K}]$</td>
<td>$3.53 \times 10^{-44}$</td>
</tr>
<tr>
<td>$U_i [\text{eV}]$</td>
<td>15.6</td>
</tr>
<tr>
<td>$K (4.65 \text{ eV} @ 266 \text{ nm})$</td>
<td>4</td>
</tr>
<tr>
<td>Raman</td>
<td></td>
</tr>
<tr>
<td>$B_e \text{ [cm}^{-1}]$</td>
<td></td>
</tr>
<tr>
<td>$f$</td>
<td></td>
</tr>
<tr>
<td>$\omega_r \text{ [}\times 10^{12} \text{ s}^{-1}]$</td>
<td></td>
</tr>
</tbody>
</table>
LIST OF REFERENCES


