Toward Development of X-ray Lasers in
“Water Window”

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Dedicated to my wife, our parents, on both sides of the Pacific Ocean
ABSTRACT

Extensive experimental efforts have been spent on the development toward lasing to the ground state in CVI ions at 3.37 nm, which lies well within the ‘water window’. The propagation of 100 fs pulses, with an intensity of $4 \times 10^{16}$ W/cm$^2$, in optically ionized gases was studied both experimentally and numerically. Ionization-induced refraction was identified to be responsible for the much shorter propagation distance in N$_2$ than in H$_2$. Three dimensional particle-in-cell simulations also showed the roles played by the forward Raman and ionization scattering instabilities in further affecting the propagation. A modified igniter-heater technique, which involved the combination of a 200 ps Bessel beam and a 10 ns spherical beam, was successfully developed in creating plasma waveguides with 450 $\mu$m diameter and over $10^{19}$ cm$^{-3}$ axial density, ideal for Raman amplification. Plasma waveguides with much smaller diameters ($<10$ $\mu$m), were produced using the combination of two Bessel beams of 100 fs and 200 ps duration, aiming at guiding the ultrashort and ultraintense pump pulses in soft x-ray lasers. Spectroscopic study was performed for the plasmas created by a 100 fs, $>10^{19}$ W/cm$^2$ pulse in an ethane gas jet. A series of population inversions in CVI ions, including 3-2, 4-2, 5-2 and 5-4, were identified, especially when the level of pre-pulse was effectively suppressed by two RG850 saturable absorbers. Preliminary experiments were conducted to demonstrate the guiding of a 25 ps pulse in plasma waveguides over many Rayleigh lengths, opening the possibility of extending the plasmas having favorable conditions and thus increasing the gain to measurable level.
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Chapter 1

Introduction

1.1 Toward Shorter Wavelengths

Ever since the first demonstration of a ruby laser at 694.3 nm in 1960 [1.1], laser scientists have continued their efforts to extend the spectral range in which lasers are operating in order to fulfill the requirement for different applications. Half a century later, major commercially available lasers can now provide us with reliable sources of radiation from ultraviolet to mid-infrared, as shown in Figure 1-1[1.2].

![Figure 1-1, List of major commercially available lasers in the spectral range from ultraviolet to mid-infrared](image)

In the direction of lengthening the wavelength, CO₂ lasers (10.6 μm) leave a clear footprint in the mid-infrared segment. Neodymium-based materials, including Nd:YLF (1.047/1.053 μm), Nd: YAG (1.064 μm) and Nd:Glass (1.06 μm), serve as the most popular working media for lasers running around 1 μm. Ti: Sapphire lasers (690-960 nm),
which play an indispensable role in the still flourishing area of ultra-short and ultra-intense optics, link up the near-infrared and visible range. On the other hand, advances in pushing the spectral range to shorter wavelengths are largely a result of the invention of excimer lasers. Representative excimer lasers, such as XeF (351 nm), KrF (248 nm), ArF (193 nm) and F₂ (157 nm), are able extend the region down to the deep ultraviolet.

Aside from these fundamental wavelengths, a series of nonlinear processes are also employed to generate beams with frequencies that are not directly accessible from lasers. Frequency doubling, or second harmonic generation, as the first nonlinear process observed after the invention of laser [1.3], is probably one of the most widely used techniques in creating new frequencies. A popular example is to double the frequency of the Nd-based 1064 nm lasers into green (532 nm) beams to pump the Ti: Sapphire crystal. Similarly, 355 nm UV light can be produced by mixing the 1064 nm Nd: YAG laser with its frequency-doubled light at 532 nm through sum frequency generation. Also commercially available is the Optical Parametric Oscillator or Amplifier (OPO/OPA), which employs the process of parametric oscillation and amplification. NKT Photonics [1.4] recently released the Argos high power OPO systems that can deliver up to 5 W beams between 1.4 and 4.6 µm in CW mode. Other processes that are of active investigation include Raman conversion and supercontinuum generation. Raman laser operating in deep ultraviolet at 275.7 nm was demonstrated with diamond as the nonlinear media [1.5]. A continuous output of 49.8 W spanning from 500 nm to 1700 nm is now accessible in all-fiber supercontinuum sources with pretty good beam quality [1.6].

Despite all of these efforts, there is still a blank region called Extreme Ultraviolet (EUV) and soft x-rays as shown in Figure 1-2 in the current laser spectrum. This region
approximately covers the wavelength range from 100 nm down to less than 1 nm, corresponding to photon energies between 10 eV up to more than 1000 eV. One of the key features of XUV and soft x-ray region is the presence of primary atomic resonances and absorption edges of most low and intermediate Z elements. For example, the K-absorption edge of Be (Z=4) is 112 eV, and the L-absorption edge of Cu (Z=29) is 933 eV, both of which fall into the region of XUV and soft-x-rays. This results in the extremely short penetration length, usually less than 1 µm, in almost all materials and thus historically inhibited the pursuit and exploration in this region.

Figure 1-2, *The wavelength and corresponding photon energy in the short wavelength region of electromagnetic radiation* [1.7]

It is also because of those primary resonances and absorption edges that radiation in XUV and soft-x-rays is extremely sensitive to materials it interacts with, both elementally and chemically. Thus a bright scientific and commercial prospect can be foreseen in developing elemental and chemical identification techniques. Also, due to its relatively short wavelengths, it is capable of seeing small structures and writing small patterns, creating new opportunities in high-resolution microscopy and lithography. Combining coherence with short pulse duration, we can also open new frontiers in the study of the structures and dynamics of matters.
Currently, there are four major directions toward developing coherent radiation sources of short wavelengths: (1) Synchrotron-based radiation (2) X-ray free electron lasers (X-FEL) (3) High Harmonic Generation (HHG) and (4) Soft-x-ray Lasers (SXL). Each of these methods has made encouraging progress but at the same time also has certain limitations.

Synchrotron sources, initially for studying particle physics, attract scientists with its broad tunability and high average power. They emit X-rays from circulating bunches of energetic (~GeV) electrons that are deflected by the magnetic field in accelerator storage rings. But the spontaneous nature of the radiation prevents it from reaching high spectral brightness and thus severely limits its applications.

![Figure 1-3, Schematic of the first FEL user facility FLASH in Hamburg, Germany](image)

Important progress was made when undulators were combined with synchrotron sources and a brand new type of photon source—Free Electron Laser (FEL)—came to the attention of scientists. The first FEL, operating at 12 µm, was developed by John Madey in 1971 [1.8]. It played only a marginal role in comparison with conventional lasers until its potential in achieving exceedingly powerful radiation in the X-ray regime was predicted more than 20 years ago and fulfilled just recently as X-FEL. In an FEL, the accelerator provides bunched relativistic electrons which radiate synchronously in the undulator due to the self-organization process on the scale of the light wavelength. The radiation power is proportional to the square of the number of electrons, resulting in a fast
scale to high peak brightness and good degree of coherence. The first VUV and soft-x-ray FEL user facility FLASH, with schematic shown in Figure 1-3 [1.9], is now able to provide pulses of 6.8-47 nm, 10-70 fs, with peak brightness of $10^{29}-10^{30}$ Photons s$^{-1}$ mrad$^{-2}$ mm$^{-2}$ 0.1% bandwidth$^{-1}$ [1.10]. In 2009, Linac Coherent Light Source (LCLS) at Stanford became the first hard X-ray FEL, delivering ultrashort coherent pulses at 0.15 nm. However, these large-scale accelerator-based FELs are not practical for table-top applications and the associated high demand on space and cost also inhibits extensive use in small laboratories. The four user facilities in the world—FLASH in Germany, VUV FEL as prototype for the Spring-8 Compact SASE Source (SCSS) in Japan, LCLS at Stanford in the USA and Fermi@Elettra in Italy—can hardly handle the beam time demand from all over the world. On LCLS, for example, only a third of the received proposals were granted beam time and typically only five 12-hour shifts were allocated to each group. This creates a significant obstacle for research groups to effectively run projects that largely rely on X-FEL beams.

In comparison, High Harmonic Generation (HHG), which basically only requires a commercially available laser focusing on a target, is of table-top dimension so it is much less costly and can be utilized by many university-scale laboratories. Rare gases were predominantly employed as the target [1.11], but encouraging results were also achieved in solid target experiments [1.12]. Physical pictures of these two types of experiments are very different (see Figure 1-4). In rare gases, electrons first tunnel free from the binding potential, which is suppressed by the electric field of the laser. Then they are accelerated in the first half cycle of the laser field and reversed in the second half cycle, returning to the vicinity of the atom. Through recombination or scattering, the electrons
undergo strong short duration (much less than optical cycle) of deceleration, leading to the emission of radiation at high photon energies. As for the solid target, the harmonics are generated at the plasma-vacuum interface that is driven to oscillate at the same frequency of the ionizing pulse by laser electric field and the ponderomotive force. The oscillating interface, due to its near-solid-state density (higher than the critical density of the laser), reflects the incident pulse and at the same time introduces a phase modulation that exhibits sidebands at multiples of the modulation frequency in the spectrum. Since the frequency of the interface oscillation is the same as the laser frequency, these modulation sidebands would represent optical harmonics of the fundamental optical frequency [1.15].

![Diagram](image)

Figure 1-4. Principles of high harmonic generation in (a) gases and (b) solids [1.13][1.14]

Output at 2.7 nm (~460 eV) was achieved in HHG using Helium as the target [1.16], but there were only several hundred photons per harmonic peak per pulse, rendering its application in microscopy and lithography impractical. Even later on with the development of phase matching [1.11] and quasi-phase matching [1.17] techniques, the conversion efficiency was still very low, usually on the order of 10^-7 in the range of 10-40
eV and $10^{-8}$ in the 40-150 eV (31-8.2 nm) range. The resulted photon flux is only marginally utilizable in microscopy and is far below the threshold for nonlinear phenomena study. Nevertheless, high harmonic pulses play a critical role in attosecond science, as well as in seeding soft X-ray lasers that are described later in this chapter.

![Penetration distances in water and protein for electrons and X-rays. Edges of “water windows” are marked by two vertical red dashed lines](image)

**Figure 1-5:** Penetration distances in water and protein for electrons and X-rays. Edges of “water windows” are marked by two vertical red dashed lines [1.20].

Soft X-ray lasers (SXL), after more than 25 years’ efforts since its first experimental demonstrations at Princeton [1.18] and Livermore [1.19] in 1985, are now able to provide reliable beams between 10 nm and 50 nm with excellent beam quality. These compact SXLs are currently the most practical tools for high resolution microscopy, micro-holography, high plasma density measurement, applications to semiconductor surface studies and nano-lithography. A lot of efforts are being put into pushing the operation wavelength of SXL further down to below 10 nm and even to the “water window”, which
is between the carbon and oxygen K-absorption edges (2.2-4.4 nm, see Figure 1-5). Radiation in the “water window” has a very distinct penetration length in water from that in proteins, which are the major components in biological specimen. Therefore, a soft x-ray microscope in this region would have high resolution ($\propto \lambda$, the wavelength) and high contrast. The superior contrast can also be revealed by a comparison of penetration depth with electrons at energies that are used in electron microscopy.

Before going into any details, some key features of SXLs are described here to illustrate its distinctiveness from conventional lasers and the consequent challenges faced by SXL scientists. First of all, lasing transitions of SXLs can only be found in ions with sufficiently high charge state, due to the required large energy intervals between upper and lower lasing levels. Unlike conventional lasers using solids or gases as working materials, desired transitions of SXLs can only be found in plasmas, which are extremely volatile, aggravated by a variety of nonlinear or even relativistic processes. Thus, a thorough understanding of the behaviors of plasmas, particularly laser-created plasmas, becomes critical in utilizing them to achieve lasing in the soft x-ray region.

Also, lifetimes of the upper levels for lasing are usually very short, typically measured in picoseconds, so the pumping energy must be delivered within this time-scale or much shorter when electron collisional de-excitations are faster than their radiative counterparts, in order to build population inversions. Such fast energy delivery, in current technology, can be realized with ultra-short laser pulses, or very fast discharges in some cases. With the development of the CPA technique [1.21], as well as the recent advances in the innovative Raman amplification [1.22], we now have much better prospect for optimizing the pumping schemes of SXLs.
A third feature of soft x-ray lasers originates from the lack of high reflectivity optics at normal incidence. Practically, there is very little reflection from optical surfaces under normal incidence because most of the incident soft x-rays will be absorbed. Wavelengths of soft x-rays are also too long to make use of the Bragg reflection on natural crystals. Although people have designed a variety of multilayer mirrors, the near-normal-incidence reflectivity in “water window” is still hovering between 15% and 20% [1.23], which makes a resonant cavity impractical. Even though we manage to reflect 100% soft x-rays, we would still need to put these optics far away from the plasmas to avoid any damage. The resulted roundtrip time will probably be longer than the lifetime of the gain, so signals cannot be further amplified after a very limited number of passes. Therefore, Soft X-ray lasers must have a sufficiently large gain-length product (GL) in order to produce reasonably high energy pulses in a single pass.

Several schemes have been proposed to achieve lasing action in the soft x-ray region, including recombination scheme [1.24], collisional scheme [1.25], inner shell ionization [1.26] and photon pumping scheme [1.27]. After many years of investigation, the collisional and recombination schemes are, so far, the most successful ones [1.28], and they are be described in detail below.

1.2 Schemes of Soft X-ray Lasers

1.2.1 Collisionally Pumped Neon-like and Nickel-like Lasers

The collisional scheme of soft x-ray lasers is an extension of some of the most widely utilized visible and ultraviolet ions lasers, the cw argon ion and krypton ion lasers, in
which the laser upper levels are predominantly excited by direct electron impact collision from the ground state of the ion stage of interest. It takes advantage of ionization bottlenecks to ensure that there is a high density of ions in particular ionization state. Specifically, the laser parameters are carefully chosen to ionize the target atoms or molecules into ions of closed electron shells, i.e. ionization bottleneck, typically Neon- or Nickel-like ions, which can survive in a wide range of density and temperature, due to the large energy gap toward the next ionization state. In the most common implementation of these lasers, the population inversion is established in a quasi-cw regime, enhanced by the distinct radiative lifetimes of the energy levels that are involved in lasing. The upper lasing levels are metastable with respect to radiative decay to the ground state, and the lower lasing levels are depopulated by strong dipole-allowed transitions.

![Figure 1-6: Simplified energy level diagram for Ni-like ions in collisional scheme soft x-ray lasers, showing the 4d to 4p lasing with 3d^{10} as the ground state [1.29]](image)

Take Ni-like ions for example (Ne-like scheme is very similar), with the simplified energy diagram shown in Figure 1-6, both 3d^94d (J=0) and 3d^94p (J=1) states are excited
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states that are populated from the ground state 3d^{10} (J=0), where J is the total angular momentum. Both 3d^94d-3d^94p and 3d^94p-3d^{10} transitions are optically allowed since ΔJ=1, but 3d^94d-3d^{10} (J=0→0) transition is forbidden. Therefore, with proper electron temperature and density, the 3d^94d level is preferentially populated, and it decays much slower to ground state than the 3d^94p level does, resulting in a population inversion between these two levels.

Figure 1-7: (a): Experimental setup for the first conclusive demonstration of collision scheme soft x-ray laser in LLNL. Grazing Incident Spectrometer (GIS) is placed on both axial and off-axis directions to compare the spontaneous emission with amplified stimulated emission (ASE), Transmission Grating Spectrometer (TGSS) is to monitor the temporal evolution of soft x-ray signal; (b): Integrated line strength of J=2 to J=1 transitions at 20.6 nm and 20.9 nm versus amplifier length [1.19]

The first conclusive demonstration of collisional scheme soft x-ray lasers was accomplished at Lawrence Livermore National Laboratory (LLNL) in 1985 [1.19], at the wavelength of 20.63 nm and 20.96 nm (J=2→1) in the Ne-like Selenium plasmas. As shown in Figure 1-7 (a), the Novette laser-target irradiation facility, mainly used for Inertial Confinement Fusion (ICF), was line-focused on a target composed of a 75 nm layer of selenium, vapor deposited on one side of a 150 nm thick Formvar substrate. The most persuasive evidence of lasing action is shown in Figure 1-7 (b), in which intensities of the 20.63 nm and 20.96 nm line are growing exponentially versus the amplifier length.
Curve fitting of this data yields a gain coefficient of 5.5±1.0 cm\(^{-1}\) and the exponential behavior of line intensities indicates that the amplifier is below saturation. The maximum energy of each lasing line didn’t exceed 0.1 \(\mu J\), with an input energy of over 1 kJ from the Novette laser, at a repetition rate of 1 to 2 shots per day.

Soon after the first demonstration, saturated operation of lasing via the collisional scheme, which typically required a gain-length product \(GL \geq 14\) [1.30], was first achieved also at LLNL [1.31], where the Nova facility delivered 2.5 kJ energy at 0.53 \(\mu m\) to pump the Ne-like Selenium ions. With a target length of 4 cm, the 20.63 nm line reached saturation after with a GL of 16. Even a higher GL (~20) was achieved at the same facility in Ne-like yttrium at a wavelength of 15.5 nm [1.32]. The total output energy of 7 mJ was also the world’s highest energy soft x-ray laser, although kilojoules of energy were used for pumping. Saturation has also been observed on the \(J=2 \rightarrow 1\) transitions in Ne-like germanium [1.33] and selenium [1.34], and on the \(J=1 \rightarrow 0\) transitions in germanium [1.35], all of which were driven by hitherto the largest optical laser facilities in the world.

Because of the rapid increase of the required pumping power, the Ne-like scheme becomes very difficult to scale to wavelengths shorter than 10 nm. Instead, lasing in Ni-like ions, first proposed by S. Maxon in 1985 [1.36], is more favorable in this respect due to its higher quantum efficiency (the laser transition energy/the excitation energy). A milestone achievement was made in 1997 by a British group [1.37], who for the first time observed saturated operation at 7.3 nm in Ni-like samarium. One of the key features of this experiment was that a low intensity (~10-30% of the total energy of all involved pulses) laser pulse was used to create a pre-plasma of low ionization stage that expanded
for more than 2 ns into a larger, more uniform gain region before the arrival of the other three beams with 75 J total energy. In standard off-axis focus geometry, these three pumping beams produced a second line-plasma with opposite density gradient. X-ray beams at 7.3 nm have been monitored with output energy of 0.3 mJ in 50 ps. Saturation intensity was reached at a GL of around 16. The same experimental setup also produced gain saturation in Ni-like Ag at a wavelength of 13.9 nm [1.38].

Further reduction of soft x-ray laser’s size, from fusion-scale facilities to a few optical tables, and further increase of the repetition rate, from a few pulses per hour to multiple Hertz, was realized by the advance of a few novel approaches, including fast discharge excitation of capillary plasmas, transient electron collisional excitation, grazing incidence pumping and high harmonic seeding.

Capillary discharges have been widely studied as sources of soft x-ray radiation for spectroscopy, x-ray lithography and microscopy. A new chapter was initiated by Rocca and his colleagues when they achieved saturated output of 30 µJ at 46.9 nm in Ne-like Ar using fast Z-pinch plasma in capillary discharges [1.40]. The gain media was created in polyacetal capillaries, 4 mm wide filled with Ar, by 39 kA current pulses of 70 ns long. A remarkable feature was its compactness: only 0.4×1 m² was required on top of an optical table, comparable to most commercially available lasers in visible or near infrared region. Excellent coherence was verified by the clear diffraction pattern from the interaction between the output soft x-ray pulse and a knife edge [1.41]. Continuing efforts in this direction increased the output power to approximately 1 mW at a repetition rate of 7 Hz by replacing the polyacetal with Al₂O₃ as capillary materials in order to decrease the rate of wall ablation and boost the heat conductivity allowing for larger
dissipation of high average discharge powers [1.42]. Also, as much as 1 mJ per pulse of good reproducibility was reached by extending the capillary lengths to more than twice the saturation length [1.43]. These successful demonstrations brought us into the real table-top regime of soft x-ray lasers and made possible wide applications in laboratories. Currently one of the main challenges in discharge-pumped lasers is to reach much shorter wavelengths (10–20 nm), in which satisfactory scaling of gain-length product is yet to be realized.

Coming back to laser-driven schemes, there has been a general trend to switch to high power pumping using drivers in picosecond range, a time-scale during which large transient population inversions can be achieved in the so-called transient gain scheme first proposed by Shlyaptsev et al. in 1993 [1.44]. It is different from the previous quasi-steady-state inversion scheme, in the sense that the short-lived population inversion is pumped directly from the ground state until collisions redistribute the populations. In other words, it is the difference of the level excitation rates, instead of radiative decay rates, that creates the population inversion.

Features of the transient scheme, including high gain coefficients, short gain duration and relatively low pumping energy, were clearly demonstrated in 1997, when lasing at 32.6 nm in Ne-like Ti (3p-3s, J=1→0) was achieved at Max Born Institute [1.45]. A 1.5 ns pulse with 4-6 J energy was first to prepare the active ions TiXIII, 1.5 ns prior to the arrival of a 700 ps, 1.5 J pulse that effectively heated up the electrons by more than 0.5 keV for effective excitation. Gain coefficient $g=19\pm1.4\,\text{cm}^{-1}$ of approximately 20 ps duration—the shortest duration by that time—was reported. The transient scheme was soon applied in Ni-like ion sequences for the Pd 4d-4p line at 14.7 nm by the same group.
at LLNL [1.46]. However, short gain lifetime and its associated gain reduction at longer target lengths became a major obstacle against gain saturation in the transient scheme. It was overcome by the traveling scheme that was realized by a simple reflection echelon. Saturated lasing from 20.3 nm to 13.9 nm, corresponding to 4d-4p transition from Nb to Ag, were observed in experimental setup of table-top sizes with the combination of a 1.2±0.5 J, 600 ps pulse followed by a 5.0±0.3 J, 1 ps pulse 700 ps later [1.47].

![Diagram](image)

**Figure 1-8:** (a): Experimental setup for the first demonstration of GRIP scheme [1.49]; (b): A closer view of the setup near the target [1.29]

The pumping energy required to achieve gain saturation in the 10-30 nm region was further decreased by the Grazing Incident Pumping (GRIP) scheme, first proposed by V. N. Shlyaptsev in 2003 [1.48]. GRIP carried forward the two-pulse technique in conventional transient gain scheme but with a key innovation in the incident direction of the pump pulse which created population inversion. Instead of being perpendicular to the gain direction (transverse pumping) as in the transient gain scheme, the main pulse in GRIP is in grazing incidence geometry (as can be seen from its name) such that the pump beam can be redirected along a selected high electron density region where soft-x-rays can be optimally amplified. The incident angle $\alpha$ is obtained from approximate relation $\alpha=(n_{e0}/n_{ec})^{1/2}$, where $n_{e0}$ is the maximum density within the gain region and $n_{ec}$ is the critical density for the main pulse. This near-longitudinal pumping significantly
improves the efficiency, as can be seen from its first experimental demonstration in 2003 with the setup shown in Figure 1-8 [1.49]. Only 150 mJ total pump energy (80 mJ for the 200 ps pulse and 70 mJ for the ps pulse) was used to achieve lasing at 18.9 nm in Ni-like Mo ions at a repetition rate of 10 Hz. Gain saturation was soon realized in a series of Ni-like ions in the $4d^1S_0-4p^1P_1$ transitions, from 16.5 nm in Ru down to 13.9 nm in Ag, with 1 J pumping energy at a repetition rate of 5 Hz [1.50].

![Figure 1-9: A typical schematic drawing of the HHG-seeded soft x-ray laser which mainly consists of a soft x-ray seed from HHG and an amplifier. Longitudinal pumping in the amplifier is employed here and it can be replaced by the GRIP scheme for better efficiency [1.52]](image)

With the considerable development in advanced pumping schemes and HHG described before, the extension of a conventional laser chain into the soft x-ray region became possible. HHG resembles the conventional oscillator to deliver the seed pulse of high optical quality, which is then amplified while maintaining the beam quality in the energetic soft x-ray amplifiers. Although the first experiment in a Ne-like Ga plasma amplifier demonstrated an amplification by a factor of only 3 [1.51], Ph. Zeitoun and his colleagues overcame the strong refraction of solid target plasmas by using Optical Field
Ionization (OFI) in gas target to construct the amplifier [1.52], with the setup shown in Figure 1-9. The 25th harmonic of an infrared laser, in close match with the $4d_{f=0} - 3d^9 4p_{f=1}$ transition in Kr$^{8+}$ ions at 32.6 nm, was amplified to gain saturation after 1.7 mm. This success largely reignited the passion for seed-injection soft x-ray lasers. Rocca’s group at Colorado State University promptly introduced the GRIP scheme into SXL experiments in their laboratory and achieved gain saturation in a series of Ne-like and Ni-like ions, including 32.6 nm in Ne-like Ti which was the first HHG seeded SXL using solid target [1.53], 18.9 nm and 13.9 nm in Ni-like Mo and Ag respectively [1.54]. More recent investigation using the variable path difference interferometer confirmed experimentally that output pulses from the above experiments already approached the Fourier limit in the spectral-temporal domain [1.55]-[1.56].

1.2.2 Recombination Soft X-ray Lasers

The idea of the recombination soft x-ray laser can be traced back to the mid-1960s, when Gudezenko and Shelepin [1.24] proposed to push the laser wavelength to the soft x-ray region in highly ionized H-like ions through three-body recombination, or collisional recombination. Three-body recombination, the inverse process of impact ionization, involves the interaction of two electrons with an ion $A^Z$ where $Z$ is the charge of the ion nuclear (e.g. C$^{6+}$ designates the fully ionized carbon) and results in the generation of $A^{Z-1}$ ion, with the other electron carrying away the excess energy:

$$A^Z + e + e \rightarrow A^{Z-1} + e$$  \hspace{1cm} (1-1)

Its rate $R_{3b}$ is very sensitive to electron density $n_e$ and temperature $T_e$ as:
\[ R_{3b} \propto n_e^2, R_{3b} \propto T_e^{-9/2} \]  

(1-2)

Therefore, high density and low temperature are favorable conditions for three-body recombination to dominate. Moreover, three-body recombination has a strong preference to populate highly excited levels in \( A^{Z-1} \) indicated by \( R_{3b} \propto n^4 \), where \( n \) is the principal quantum number of \( A^{Z-1} \) ions.

![Diagram](image)

**Figure 1-10:** Simplified energy level diagram for H-like ions in recombination scheme soft x-ray lasers, showing the 3 to 2 lasing in CVI ions [1.57].

The physical picture of the recombination scheme can be described using the lasing at 3-2 transition in CVI ions (H-like carbon, or five-time-ionized carbon \( C^{5+} \)) as an example, with the energy diagram shown in Figure 1-10. The carbon atoms have to be totally stripped of electrons first, followed by three-body recombination that dominantly populates the highly excited levels. The electron impact de-excitation, whose rate drops as the energy gap between the two involved states increases (\( R_{\text{ed}} \propto (\Delta E)^{-1/2} \)), rapidly...
transfers the population to lower levels until a bottleneck level (n=3 here) is reached when impact de-excitation is outran by the radiative decay. In this case, impact de-excitation keeps populating level 3, whereas level 2 is quickly losing its population to the ground state through radiative decay. The 3-2 population inversion is thus established.

![Figure 1-11: Scaling of the wavelength versus ion charge for (a) both Ni-like and Ne-like ions, in collisional scheme, and (b) H-like ions for 3-2 and 2-1 transitions in recombination scheme [1.29].](image)

Lasing transitions in the recombination scheme occur between different principal quantum levels so their scaling to short wavelengths with respect to Z is quite fast, especially for lasing to ground states, as can be seen in Figure 1-11. 13.5 nm can be reached in LiIII (Z=3) ions and 3.37 nm in CVI (Z=6) ions with only moderate Z. As a comparison, a much higher Z is required in order to attain similar wavelengths in the collisional scheme (e.g. 13.9 nm in Ni-like Ag, Z=19). So, the recombination scheme is very attractive in reaching the “water window” using currently available multi-terawatts systems of compact size at a convenient repetition rate (5 Hz or more).
However, plasma conditions favoring the population inversion in the recombination scheme seem to be in conflict with each other; on the one hand we need fully stripped ions but on the other hand a low plasma temperature is also necessary. Therefore, efficient cooling becomes critical and it is usually realized through the following three mechanisms or some combination of them: 1) Adiabatic expansion into vacuum, which is most effective for high density, small diameter plasmas [1.58]-[1.61]; 2) Electron conduction to colder surroundings, such as a solid wall or gases [1.62]; and 3) Radiation from high-Z ions intentionally introduced as impurities [1.18][1.63].

The first clear demonstration of recombination-type soft x-ray lasing, which was published in 1985 by S. Suckewer and his group here at Princeton [1.18], utilized a 75 ns, 300 J, CO₂ laser pulse to irradiate a solid carbon disk as shown in Figure 1-12 (a). They observed a gain-length product $GL \sim 6.5$ for the CVI 3-2 transition at 18.2 nm from comparing the soft x-ray signal in axial and transverse directions shown in Figure 1-12 (b). Another comparison was made with the 4-2 transition at 13.5 nm in CVI ions, which has the same lower level and upper level of close potential as the lasing line.

![Figure 1-12](image)

**Figure 1-12**: (a): Experimental setup for the first clear demonstration of lasing at 3-2 transition in CVI ions; (b): Time evolution of CVI 18.2 nm and 13.5 nm line intensities measured with axial and transverse XUV instruments with same plasma conditions [1.18].
Radiation cooling was believed to be dominant in this experiment while the carbon blades provided additional cooling through heat transport. A novel solenoidal magnetic field of 90 kG was used to confine the plasma column and thus maintain a relatively high plasma density. Lasing was further verified using a soft x-ray mirror, the first reported effort to build cavity structure in the soft x-ray region. With only 12% reflectivity from the cavity mirrors, the intensity of the 18.2 nm line in the axial direction increased by 120% while the transverse signal of the same line stayed the same.

Lasing at 18.2 nm in CVI ions was also achieved in the VULCUN laser system at Rutherford Appleton Laboratory using 70 ps [1.61] and 2 ps [1.64] pulses respectively. Other wavelengths, including 8.1 nm in FIX and 5.42 nm in NaXI, were reported to be amplified before 1990 [1.59]-[1.60]. Conduction cooling [1.62] and radiation from highly ionized Selenium impurities [1.63] were also shown to be effective in achieving lasing in this CVI Balmer-α line. Although the original proposal only considered H-like ions as the lasing medium, Li-like ions also proved to be practical and gain was observed in Li-like Al from several groups [1.65]. Lasing on 3-2 transition of BV ions at 26.2 nm was even realized in a compact table-top system with 1 Hz repetition rate [1.66].

As mentioned before, lasing to ground states is particular advantageous in scaling to shorter wavelengths. Although the theoretical proposal was first published in 1975 [1.67], no practical method was elaborated by that time. The key issue was still cooling, whose time scale had to be at least shorter than the lifetime of the upper level involved in lasing in order to avoid the over-population of ground states, which would easily kill the gain. For instance, the cooling has to be completed within 26 ps and 1.6 ps, lifetimes of n=2 in LiIII and CVI ions respectively, in order to achieve lasing to their ground states. For
quite a long period of time this approach was ruled out from experiments that relied on the previously mentioned cooling methods, which usually takes a longer time than that is required, until the advent of powerful subpicosecond laser systems [1.21] and detailed theories of creating cold dense plasma through Optical Field-Induced (OFI) ionization [1.68].

![Diagram of experimental setup](image)

**Figure 1-13**: (a) Experimental setup for creation of plasma in LiF microcapillary with Nd/glass laser and optical field ionization with 250 fs, 248 nm Excimer laser; (b): Line intensity of 13.5 nm and 11.4 nm in LiIII ions as a function of microcapillary length [1.73].

In 1989, predictions of cold plasma creation using linearly polarized ultrashort high-intensity laser pulses were made by Burnett and Corkum, who calculated that the electrons stripped through OFI ionization can be much colder than the ionization potential of the parent ions [1.68]. OFI ionization occurs when a single photon has much lower energy than the ionization potential ($h\nu<<E_i$). The electrons are stripped either by the simultaneous absorption of multiple photons (multiphoton ionization) in the weak
field limit, or by tunneling through the Coulomb barrier that is temporarily suppressed by the strong laser field (Above-Threshold Ionization or ATI), which is usually above $10^{14}$ W/cm$^2$. In the latter case, the tunneling rate is strongly dependent on the laser intensity and can be tuned to dramatically outrun the radiative decay of the upper lasing level. Also, electrons are compelled to return their quiver energy back to the laser field in linearly polarized pulses. Therefore, fully stripped ions and low plasma temperature can be simultaneously achieved. Since the inverted population is a result of different collisional de-excitation rate from upper levels before radiative decay sets in, this scheme is also called the collision-dominated transient scheme, instead of the quasi-steady state collisional-radiative scheme used for lasing at 3-2 transitions before.

This approach was first experimentally realized in 1993 at RIKEN [1.69], where a nonlinear increase of the 13.5 nm line (2-1 transition in LiIII ions) versus plasma length up to 2 mm was observed. However, the failure to observe further intensity increase of the 13.5 nm line with the plasma length made uncertain the gain validation of 20 cm$^{-1}$. The first unequivocal demonstration came in 1996 [1.73], when Korobkin et al. at Princeton increased the amplification length to 5 mm using plasma waveguide technique as illustrated in Figure 1-13 (a). The low power (100 mJ, 5 ns) Nd:YAG laser ablated the microcapillary wall and created pre-formed plasma, which evolved into a hollow-pipe structure with minimum density on the axis when the powerful subpicosecond laser (50 mJ, 250 fs, $10^{17}$ W/cm$^2$ at focus) arrived hundreds of ns later. This hollow-pipe density profile corresponded to a refractive index distribution resembling a positive lens. It guided the propagation of the powerful ionizing pulse, as well as the soft x-ray signals along the microcapillary axis. The observed exponential increase of the 13.5 nm line of
H-like Li versus length is shown in Figure 1-13 (b), and a gain-length product $gL \approx 5.5$ was derived. Besides H-like ions, OFI-pumped lasing in plasmas of low-charge ions, mostly Oxygen and Nitrogen, were also reported [1.71]-[1.74], aided by their close-to-unity ratio of statistical weights between the two levels involved in lasing (favorable for population inversion).

In parallel with experimental efforts, computational investigation becomes increasingly necessary to identify the physical processes involved in gain creation and to optimize the methods in producing suitable conditions for lasing. Out of the numerous simulation results regarding the lasing to ground states, the most encouraging one came from the elaborate model built by Avitzour [1.75], who first took into account the non-Maxwellian nature of electron energy distribution immediately following the OFI ionization, as shown in Figure 1-14 (a). The importance of non-Maxwellian distribution with excess electrons in the low energy region, and the subsequent enhancement of the three-body recombination rates, was recognized as early as in 1996 by Ditmire [1.78]. However, it was not utilized in calculating the rates of atomic processes that participated in building up the population inversion until in 2004, when Avitzour integrated this feature into his model and successfully explained the high gain observed in experiments in LiIII ions [1.76]. A series of experimentally-adjustable parameters, including lasant ion densities, spot size, pumping beam wavelength and peak intensities, was also investigated in detail for optimal design of soft x-ray lasers.

The doping with atoms of low ionization potentials, such as hydrogen, was another important step in further reducing the plasma electron temperature and thus constructing practical soft x-ray lasers based on OFI. Electrons acquire most of their energy from
residual heating, which is proportional to the instantaneous intensity of the pulse at the moment when the ionization occurs, therefore, hydrogen atoms, which will be ionized at the very beginning of the pulse, can provide a high-density cold-electron environment. Grout et al. first considered the H₂/N₂ and H₂/Ar mixture as the gas target for recombination-scheme soft x-ray lasers [1.79], followed by a series of numerical works [1.80], all of which confirmed a significant enhancement of gain by the introduction of hydrogen. This method was also included in the numerical model designed for lasing to ground state in CVI at 3.4 nm [1.75]. Combining with the non-Maxwellian distribution, a very high gain of 210 cm⁻¹ was predicted as shown in Figure 1-14 (b) using a 400 nm, 50 fs pulse focused into a diameter of 15 μm in a gas mixture with 1:10 carbon:hydrogen initial density ratio, a very practical condition that can be realized with current technologies.

![Figure 1-14: Calculated results for carbon density of n_c=10¹⁹ cm⁻³, hydrogen density of n_H=10²⁰ cm⁻³, pump beam diameter of d=15 μm, wavelength λ=400 nm, pulse duration τ=50 fs and peak intensity I=1×10¹⁹ W/cm². (a) Comparison of the electron energy distribution following OFI with Maxwellian distribution with the same average energy; (b) Gain, in cm⁻¹, on CVI ions with hydrogen added, versus time and space.](image-url)
Translating the high gain coefficient into large gain-length product will be the next critical issue in realizing practical soft x-ray lasers. It comes from simulations that plasmas of at least a few millimeters long are necessary to achieve gain saturation and thus maximum extraction of energy from the lasing medium. Maintaining lasing conditions over such a distance is challenging, since the pumping beam is usually of ultra-high intensity \(10^{17}-10^{19} \text{ W/cm}^2\) and its propagation in gases or underdense plasmas can hardly be longer than a few mm due to a variety of nonlinear phenomena, which are difficult to control experimentally. A few methods have been proposed and experimentally confirmed in guiding ultra-intense laser pulses, including charge-displacement channeling, relativistic self-channeling [1.84], and plasma waveguide [1.85], which are discussed in detail in Chapter 3 and Chapter 4.

1.3 Thesis Outline

Based on the introduction above, the main objective of this thesis is to examine the potential to achieve the lasing to ground state in CVI ions at 3.37 nm using as a gain medium the plasmas created by the interaction of ultrashort pulses (100 fs) with ethane gases ejected from a gas jet. A 3-2 lasing at 18.2 nm is also investigated using a 25 ps pulse. Special attention is paid on the plasma waveguide method for the propagation of powerful laser pulses, a critical step to enable the creation of extended gain region and thus the possibility of gain measurement. These tasks are incorporated into the following five chapters.
Chapter 2 describes the laser systems that are involved in our investigation, including a multi-terawatts Thales Alpha laser system and the less powerful—but still in terawatt regime—Positive Light system.

Chapter 3 studies the propagation of ultrashort pulses in gases and optically ionized plasmas, identifying the role of ionization induced refraction, forward Raman scattering and ionization scattering as disrupting factors.

In Chapter 4 we perform extensive experiments to develop plasma waveguides using a modified igniter-heater technique. Two types of plasma waveguides are created, not only for soft x-ray lasers, but for the Raman amplification as well.

Chapter 5 is about the spectroscopic study of the plasma created by ultrashort pulses in a gas jet. Plasma conditions, especially the ion species, relative level population etc, are presented together with its prospect for gain generation.

Chapter 6 summarizes the work in this thesis and provides thoughts, suggestions and directions for future work.
References:


Chapter 1. Introduction


Chapter 1. Introduction


Chapter 2

Multi-Terawatts Laser Systems

2.1 Overview

Two laser systems operating at terawatt level—Positive Light and Thales Alpha—are the major experimental tools for most of our experiments related to the development of soft x-ray lasers. They are both based on the Chirped Pulse Amplification (CPA) technique and share the same Ti: Sapphire front end, which delivers positively chirped seed pulses of 1-2 mJ and 200 ps. This chirped pulse is then split into two parts to seed the Positive Light (single stage four-pass amplification) and Thales Alpha (two stages of four-pass amplification) amplifiers with outputs of 700 mJ and 2 J per pulse respectively. A grating compressor is installed after each amplifier to recompress the pulses into femtosecond regime by compensating the chirps introduced by the stretcher before the regenerative amplifier. Positive Light system has a final output of 400 mJ at 200 fs, translating to a power of 2 TW. An energy splitter that is made of wave plates and polarizers is installed between the final amplifier and the compressor in the Thales Alpha laser in order to simultaneously deliver pulses of 200 ps and 100 fs, with adjustable energies for each pulse in order to satisfy requirements for various experiments. At a repetition rate of 5 Hz, the 200 ps pulse has a maximal energy of 2 J per pulse, and the 100 fs output is up to 1.4 J per pulse, giving a power of 14 TW. The block diagram of the whole system is schematically shown in Figure 2-1, and detailed description is presented in this chapter.
Chapter 2. Multi-Terawatts Laser System

2.2 Front end

The discovery of Ti: Sapphire crystal as a laser medium is probably one of the most significant advances in ultra-fast optics. Its advantage in producing and amplifying ultrashort laser pulses is established by two unique features. First, Ti: Sapphire crystal has an ultra-wide gain bandwidth from 650 nm to 1100 nm [2.1], which easily satisfies the time-bandwidth product determined by the uncertainty principle ($\tau \Delta v = 0.44$ for Gaussian pulses and 0.315 for sech$^2$ shape). For example, a Gaussian pulse of 100 fs at 800 nm requires a minimal gain bandwidth of 9.4 nm. The other feature comes from its intensity dependent refractive index $n$ which induces the Kerr lens mode-locking [2.2]. A simplified expression for refractive index can be written as $n = n_0 + n_2 I$, where $n_0$ is the...
linear component and I is the laser intensity. The non-linear index $n_2$ of Ti: Sapphire crystal is typically $\sim 3.3 \times 10^{-16}$ cm$^2$/W at 550 nm and monotonically decreases to $\sim 2.8 \times 10^{-16}$ cm$^2$/W at 1550 nm [2.3]. Therefore, shorter pulses of higher intensity would experience in the laser rod more significant self-focusing, which reduces the beam size and thus further increase its intensity. This loop of positive feedback makes the initially shorter pulses quickly outrun the longer ones and is believed to be responsible for the self-starting and self-sustaining of the mode-locking.

However, due to the broad spectrum associated with ultrashort pulses, the group velocity dispersion (GVD) in the laser rod introduces a positive frequency chirp into the pulse and lengthens the pulse duration. Negative dispersion can be generated by pair prisms [2.4] for compensation and 60 fs pulses were generated using this technique [2.5]. Further compensation for high order dispersion terms from prism pairs can be achieved by double chirped mirror (DCM) [2.6], which can decrease the pulse duration to as short as 5 fs [2.7]-[2.8].

![Schematic of the Tsunami Mode-locked Ti: Sapphire laser head](image)

*Figure 2-2, Schematic of the Tsunami Mode-locked Ti: Sapphire laser head*
The Ti: Sapphire oscillator (Tsunami) in our laboratory is manufactured by Spectra Physics, and its schematic is shown in Figure 2-2 [2.9], but without the optional Model 3930 of Lok-to-Clock electronics. The mode-locking can be achieved in either passive or active way. Passive mode-locking is the Kerr lens mode-locking as described above. An acousto-optic modulator (AOM), which is driven by regenerative RF signal, is responsible for the active mode-locking.

When the laser is in the process of alignment, it is operating in a continuous mode with oscillations from several longitudinal modes. They are partially phase-locked, and the mode beating generates a modulation in the laser output at a frequency of \( \frac{c}{2L} \), where \( c \) is the speed of light and \( L \) is the cavity length. A photodiode then picks up the beating and divide it by two in order to match the driving frequency for the AOM, which in turn introduces periodic loss to filter out pulses that are out of phase. This design removes the stringent requirement of precise cavity length matching the external drive frequency, one of the biggest drawbacks of conventional active mode-locking. Therefore, even the cavity length changes (e.g. due to thermal effect), regenerative AOM, in together with the Kerr lens effect, will self-adjust correspondingly and ensure a stable operation of the mode-locking without dropouts or shutdown.

Two prism pairs, \( \text{Pr}_1 \) to \( \text{Pr}_4 \) are employed to compensate the GVD originated mostly from the Ti: Sapphire crystal. Although they also introduce third or higher order dispersions, the advantage to continuously adjust the negative GVD is favored by most of the manufacturers. The prism pairs also spread the laser frequency spatially. By changing the position and width of the slit between the prisms, we can actually adjust the output wavelength and bandwidth correspondingly.
Early Ti: Sapphire lasers were pumped by Argon ion lasers at 488 nm/515 nm [2.5],[2.7]-[2.8]. In our system, a more stable, frequency-doubled Nd:YVO₄ laser (Millennia) at 532 nm is employed to end pump the laser rod in Tsunami. Both wavelengths of Ar⁺ and frequency-doubled Nd:YVO₄ lasers are overlapping well with the Ti: Sapphire absorption band ranging from 400 nm to 630 nm.

![Figure 2-3, Schematic of the Millennia V Diode-pumped CW laser head](image)

The schematic of the Millennia is shown in Figure 2-3 [2.10]. Emission at 1064 nm from the Nd:YVO₄ crystal is directed to the intra-cavity frequency doubling arm with a non-critically phase-matched, temperature-tuned LiB₃O₅ (LBO) crystal [2.11]. Two advantages make the LBO crystal particularly suitable for high power intra-cavity frequency doubling. First, it is transparent between 160 nm and 2.6 µm with a high damage threshold of 25 GW/cm² (at 1064 nm, 0.1 ns pulse). Second, it is robust against slight misalignment due to its large acceptance angle so LiB₃O₅ crystal can work stably
inside the cavity. A small, low-power oven is used to regulate the crystal temperature for phase-matching and maintain a stable output.

The Millennia is specified to have an output of 5 W at 532 nm in order to effectively pump the Tsunami, which, according to the specification, provides an average power of 700 mW at a repetition of 82 MHz for 100 fs pulses, with each pulse of ~8.7 nJ energy. However, the unavoidable aging effect noticeably decreases the output of Millennia laser to less than 4 W, so accordingly the pulses from Tsunami would have less energy. They will be sent to a regenerative amplifier followed by two separate multi-pass amplifiers. The amplifiers operate in saturated regime so part of the energy loss in the oscillator can be compensated during the amplification. The timing of the pulse train is used to synchronize the rest of the laser system, as well as other experimental devices, including the gas jet and CCD cameras.

2.3 Regenerative Amplifier

A regenerative amplifier is a device in which a selected laser pulse from a pulse train can be trapped in the optical resonator for multi-pass amplification before being released as output through an optical switch, usually realized by an electro-optic modulator and a polarizer. We use the Quanta-ray TSA regenerative amplifier as shown in Figure 2-4 [2.12], and its working principle can be illustrated by a three-step process.

Step 1: The 5 Hz pump beam from a Quanta-Ray laser, in synchronization with the 82 MHz RF signal, pumps the Ti: Sapphire crystal R1 and the two Pockels cells PC1 and PC2 are off (voltage is off). The S polarized input pulses, generated by stretching the output of the oscillator (100 fs, ~8.7 nJ) to 200 ps by a grating stretcher, are reflected into
the cavity by the Ti: Sapphire rod R1 that has an end cut in Brewster’s angle. The input pulses will go through the quarter-wave plate WP1 twice when being reflected back by M1 and thus become horizontally polarized. These pulses continue traveling in the cavity until another double passes through the WP1 rotate them back to vertical polarization. The polarizer P1 then reflects these pulses out of the cavity without much amplification.

![Schematics of the regenerative amplifier](image)

**Figure 2-4, Schematics of the regenerative amplifier**

**Step 2:** The trapping and amplification is initiated by turning on the PC1 when one horizontally polarized pulse (seed pulse) is traveling between PC1 and M2 after the laser rod R1 is sufficiently pumped. Now PC1 works as another quarter-wave plate and cancels out the effect from WP1, leaving the cavity essentially free of components that change the polarization of pulses. So throughout this stage, the seed pulse is bouncing back and forth in the cavity for amplification. Although successive input pulses are still being injected into the cavity, their vertical polarization determines that they will be reflected out of the cavity by the polarizer P1 within one round trip.

**Step 3:** After a certain number of passes (usually ~20), another quarter-wave voltage is applied to PC2, which, after two passes, rotates the amplified pulse by 90°. P1 then
reflects this pulse out of the cavity as output, completing one working cycle of the regenerative amplifier.

The regenerative amplifier has amplification near $10^6$, so its output pulses are of 1-2 mJ at 200 ps. These pulses are then split into two parts, as we have already indicated, and directed into two separate multi-pass amplifiers—Positive Light and Thales—after passing through a single pulse selector (part of the Positive Light system) for contrast ratio improvement.

2.4 Four-pass Amplifier System (Positive Light)

The Positive Light four-pass amplifier system is schematically shown in Figure 2-5 [2.13]. It consists of two major parts: the single pulse selector and the amplifier.

![Figure 2-5, Schematics of the Positive Light four pass amplifier](image)
2.4.1 Single Pulse selector

In ideal operation of regenerative amplifiers, only one pulse should be emitted in each working cycle. However, there are always some smaller pulses preceding or following the main pulse, with their origins mostly from the following four sources.

1) Amplified spontaneous emission (ASE). This pulse has approximately the same spectrum as the gain spectrum of the amplifier, and is usually of ns duration.

2) Finite extinction ratio of the polarizer. A small fraction of the circulating pulse, although it is horizontally polarized, is reflected out of the amplifier each time the pulse passes through P1.

3) Limited contrast ratio of the Pockels cell. A small mis-alignment, or thermal effects-induced birefringence, or a slight change of the voltage in the Pockels cell, will introduce a small vertical polarization component to the pulse that will then be reflected by P1. The above two types of leakage generate a sequence of premature pulses separated by the round-trip time of the regenerative amplifier.

4) Unequal cavity lengths between the oscillator and the regenerative amplifier. Ideally, a new pulse arrives as one pulse is leaving the resonator. But with unequal cavity lengths, there are always two pulses in the resonator separated by the difference in round-trip time. If this extra pulse is approaching the Pockels cell during the switching, a small fraction of this pulse will also stay in the resonator and be amplified.

These noise pulses, although of relatively low energy, can acquire considerable amplification in the amplifiers and introduce usually unwanted effects in the experiments,
e.g. creating pre-plasmas that will disrupt the propagation of the main pulse in gases or underdense plasmas.

The single pulse selector, consisting of a Double Crystal Pockels Cell (DCPC) and a Glan-Taylor polarizer (Glan), is placed right after the regenerative amplifier to improve the contrast ratio (intensity ratio between main pulse and additional pulses). The Glan-Taylor polarizer is optically crossed with polarizer P1 so no pulse from the regenerative amplifier can pass through if DCPC is off. The DCPC has two quarter-wave plates which are optically in series but electrically in parallel so application of a quarter-wave voltage introduces a half-wave rotation and enable the TSA pulses go through. The onset of the DCPC is controlled by the same timing unit for the regenerative amplifier. Therefore, it can be turned on exactly when the main pulse arrives with less than 1 ns jitter.

2.4.2 Four-pass Amplifier

The pulse with improved contrast then go through a beam splitter (BS1), which directs part of the beam to Thales system while the other half enters the four-pass amplifier.

A telescope expands the pulse diameter from 2 mm to 10 mm in order to reduce the incident power on the surface of the Ti: Sapphire crystal which has a diameter of 16 mm and length of 19 mm. A ‘bow-tie’ arrangement is employed for the pulse to travel four passes through the crystal and thus receive manifold amplification.

The 5 Hz pumping beams of the crystal are from a powerful Nd: YAG laser system. The 400 mJ pulses at 1064 nm from a Quanta-Ray laser are amplified to 5 J after passing through two 9.5 mm and 15.8 mm Nd:YAG rods. A KDP crystal then converts the infrared beam into 2.5 J at 532 nm, which is split equally into two parts in order to pump
the crystal uniformly from both ends. The pumping uniformity is further improved by shaping the green beams into a flat-top profile through relay imaging systems. Two glass tubes mounted on the amplifier bench are evacuated to ~0.1 Torr to avoid air breakdown at the focus of the green beams. Under this pumping condition, the Positive Light system is capable of delivering 700 mJ, 200 ps beams at 800 nm, at a repetition rate of 5 Hz.

2.5 Thales Alpha Laser System

The reflected portion of the pulse from the beam splitter BS1 after the single pulse selector is directed into the Thales Alpha laser system, the most powerful system in our laboratory. It consists of two four-pass amplification stages pumped by the second harmonics of five Q-switched Nd: YAG lasers (Saga1 to Saga 5) as shown in Figure 2-6.

![Figure 2-6](image-url)

Figure 2-6, Schematics of the Thales Alpha system, 800 nm beams are illustrated in red and pumping beams (532 nm) are in green.
The first stage of amplification is very similar to the Positive Light system. The input pulse travels four times through the Ti: Sapphire crystal which is pumped from both ends by relay-imaged beams equally split from the output of Saga1. This stage amplifies the seed pulse from 1 mJ to approximately 35 mJ.

The second stage is pumped by four Saga lasers that are divided into two groups. Saga2 and Saga3 pump the left side of the laser rod, while the other side is pumped by Saga4 and Saga5. Each of the Saga laser has a specified output of 1 J per pulse at 532 nm at a repetition rate of 5 Hz. But their performances gradually degrade over time so we need to routinely balance their output energies in order to maintain a uniform pumping of the crystal. The diameter of the amplified pulse from the Stage1 is enlarged from 5 mm to 16 mm before entering the four passes of Stage2 to avoid damages. This stage is capable of delivering 200 ps pulses of 2 J, 800 nm at a repetition rate of 5 Hz.

![Figure 2-7, The wave plate-polarizer energy split system](image)

Between the Faraday isolator and optical compressor, we have installed a homemade energy split system to enhance our control over the laser output. The system is made of two wave plates (WP1 and WP2) and three polarizers (P0, P1 and P2) as is shown in Figure 2-7. The pulse first transmits through polarizer P0 and becomes horizontally
polarized. Then depending on rotation angle, wave plate P1 introduces to the pulse various amounts of vertically polarized components, which are able to go through polarizer P1 for experimental use. The WP2-P2 combination does the same control over the part of the pulse that is reflected by P1 and deliver adjustable amount of energy to the compressor. The portion that passes through P2 is dumped to a piece of foam for safety. Figure 2-8 shows a measurement of the energy transmitted through P1 versus the rotation angle of the wave plate WP1. With this energy splitter, the Thales Alpha system can simultaneously deliver pulses of 200 ps and 100 fs, with the energy of each pulse being independently adjustable. We can therefore construct a compact setup using only one laser system for soft x-ray laser experiments because the 200 ps pulse is capable of creating plasma waveguides that guide the propagation of the 100 fs one.

![Figure 2-8](image)

**Figure 2-8, The power transmitted through P1 as a function of the WP1 angle**
2.6 Faraday Isolator

Most of the experiments conducted in our laboratory using the powerful output from the two laser systems involve the creation of plasmas, which inevitably reflect a small portion of the incident beam back to the laser chain. This portion, although small, can be considerably amplified, if there is still some leftover of population inversions in the laser rods, and thus cause damages to optical components. The situation would become even worse if the reflected pulse is ultrashort (e.g. 100 fs as in the Thales laser), which is capable of directly damaging the Ti: Sapphire crystal during the amplification.

![Figure 2-9](image)

**Figure 2-9**, Schematics of the Faraday isolator with the arrows in rectangles indicating the polarization directions of a forward and backward-propagating beam

Our solution was to install a Faraday isolator immediately after the two powerful amplifiers. The Faraday isolator basically consists of a Faraday rotator placed between two polarizers with their optical axis crossed at 45° as shown in Figure 2-9 [2.14]. Polarization rotation $\alpha$ in the Faraday rotator is empirically given as $\alpha=VBd$, where $V$ is a constant depending on materials, $B$ is the magnetic flux density in the direction of light propagation and $d$ is the length of the Faraday rotator. An important aspect of magnetic
field-induced rotation is that the change of polarization direction is only defined by the magnetic field direction and the sign of the V constant. If a linearly polarized beam is sent through a Faraday rotator and back again after being reflected by a mirror, the polarization changes of the two passes add up, rather than canceling each other.

The magnetic field, generated by permanent magnets in our case, is chosen such that the polarization of the incoming beam will be rotated by 45°. Therefore, the output from amplifiers, usually in vertical polarization, passes through the first polarizer pol 1 and then undergoes in the Faraday rotator a clockwise (if we look along the propagating direction) quarter-wave rotation that enables it to pass through the second polarizer pol 2 without loss. However, pulses that are reflected back from plasmas will not be able to reach the other end of the system, due to the non-reciprocal nature of Faraday rotation. The reflected pulse reverses its propagating direction with respect to the magnetic field so the rotation angle will be 45° in counterclockwise direction. But also because of the reversal propagating direction, the helicity also reverses. Then this rotation converts the pulses into horizontal polarization, instead of bringing them back to vertical ones, and enables the first polarizer pol 1 cleanly dump the reflected pulses.

2.7 Stretcher and Compressor

The key element in a Chirped Pulse Amplification (CPA) system (see e.g. review paper [2.15]) is the grating stretcher-compressor pair. The stretcher is to stretch, in time domain, the ultra-short pulses from the Ti: Sapphire oscillator by introducing certain chirp to the pulse in frequency domain, in order to avoid damages to the subsequent amplifiers. After the chirped pulse acquires enough energy, the compressor, which
perfectly matches the dispersive property of the stretcher, then compensates the chirp and thus transforms the pulse, in principle, back to its original duration.

The stretcher and compressor in our system are schematically shown in Figure 2-10. The pulse enters the stretcher through the reflector M1 and reaches the grating G1 through the retro-reflector R1 that is made of two mirrors perpendicular to each other. The grating diffracts the pulse into a spherical mirror SM of focal length $f$, which introduces a positive chirp to the pulse, i.e. lower frequency components of the pulse travel shorter distances than the higher ones. Then R1 picks up the pulse after it is diffracted back to G1 and shifts its height so it can complete another round trip through the grating-SM system, making the total number of diffractions to four, in order to gain more chirp and thus larger stretching ratio. Larger number of round trips inside the stretcher would introduce larger stretching ratio but also more significant energy losses, as well as challenges in alignment, which renders such design inefficient.

Figure 2-10, Grating stretcher and compressor in the CPA system
The 200 ps, positively chirped pulse then goes through the regenerative amplifier, Positive Light or Thales Alpha system, and Faraday isolators before being re-compressed. The compressor we use consists of a pair of grating G2 and G3, a retro-reflector R2, as well as the input and output coupling mirrors M3 and M4. The group time delay from the grating pair compressor in one round trip can be written as [2.17]:

\[
\tau(\lambda_0 + \Delta \lambda) = \tau_0 + \frac{2\lambda_0 L_2}{cd^2 \cos^3 \theta_0} \Delta \lambda + \frac{3L_2}{cd^2 \cos^3 \theta_0} \left(1 + \frac{\lambda_0 \sin \theta_0}{d \cos^2 \theta_0}\right)(\Delta \lambda)^2 + O(\Delta \lambda^3)
\]  

(2-1)

where \(\lambda_0 = 800\) nm is the central wavelength of the pulse, \(L_2\) is the distance between the two gratings, \(c\) is the speed of light and \(d\) is the grooves spacings of the grating. The diffracted angle \(\theta_0\) satisfies the grating equation:

\[
d(sin(\theta_0 - \alpha_s) + sin(\theta_0)) = \lambda_0
\]  

(2-2)

Physically, the first term in the group delay formula is the overall time delay and is wavelength independent. The pulse duration is mainly determined by the second and third terms (we neglect the higher order terms here). In order to fully compensate the chirp introduced by the stretcher, the design of the compressor has to satisfy the following relations [2.17]:

\[
\alpha_s = \alpha_c; L_2 = 2(f - L_1)
\]  

(2-3)

Here, \(\alpha_s\) and \(\alpha_c\) are the diffraction angles of grating G1 and G2, \(f\) is the focal length of the spherical mirror, \(L_1\) is the distance between grating G1 and the spherical mirror in the stretcher, and \(L_2\) is the distance between the grating pair in the compressor (see Figure 2-10). In this case, the group delays introduced by the stretcher and compressor have the same magnitude in the first and second order terms, but of opposite signs so they cancel.
each other. Thus all the frequency components in the pulse after the compressor are delayed to the same extent \((i.e.\) no chirp) and the original pulse duration can be recovered. But the inevitable gain narrowing during the amplification, plus some nonlinear effects such as self-phase modulation, can distort the pulse in frequency domain and thus lengthens the pulse duration to be slightly more than 100 fs.

Two separate compressors are installed after the Faraday isolators for the powerful amplifiers. We conduct auto-correlator measurements during the alignment of the compressors to make sure that minimal pulse duration is achieved. The Positive Light system has a final output of 400 mJ of 200 fs duration, and the Thales Alpha system can deliver 100 fs pulse up to 1.4 J, both of which are in terawatts regime. The Thales Alphas system, with 14 TW output power, is our major tool in the investigation toward soft x-ray lasers at 3.4 nm and the Positive Light plays an indispensable role in the diagnostics for plasmas and creation of pre-plasma to guide the ultra-short and ultra-intense pulses.
Reference:
Chapter 3

Propagation of Ultrashort Pulses in Gas Jet

One of the key issues in recombination soft x-ray lasers is the propagation of laser beams through gases or plasmas in order to maintain the lasing conditions (population inversion) over a distance long enough to reach a significant gain-length product. For the soft x-ray laser to reach a wavelength of 3.4 nm, the pump pulse must be ultra-short ($\leq 100$ fs) and ultra-intense ($\geq 5 \times 10^{18}$ W/cm$^2$) according to the extensive numerical simulation [3.1].

Propagation of laser pulses in gases and plasmas may be affected by a variety of non-linear phenomena, including the optical Kerr effect and stimulated Raman scattering [3.2]. Therefore, a detailed study and good understanding remains relevant, although a significant number of papers have been published starting with pioneering ones [3.3]-[3.6], followed by more experimental [3.7]-[3.9] and theoretical [3.10]-[3.14] papers. In this chapter, we present both experimental and numerical investigation on the evolution of laser fields and plasma channels during the propagation of a 120 fs pulse through a gas jet, identifying the roles played by ionization-induced refraction, forward Raman scattering and ionization scattering.
3.1 Theoretical Considerations

3.1.1 Optical Field-Induced (OFI) ionization

During the interaction of atoms with laser fields, there exist two distinct mechanisms for plasma creation: electron impact ionization and optical field induced (OFI) ionization. Electron impact ionization, or avalanche ionization, requires the free electrons to have sufficient energy in order to ionize the atoms or ions upon collision, resulting in more free electrons that can go through the same cycle. This process usually dominates in interactions involving nanosecond pulses which allow enough time for inverse bremsstrahlung absorption to heat up the electrons. In our experiment, when the pulses are ultra-short and ultra-intense (~100-120 fs), the dominant mechanism is OFI ionization, which can be further divided into multi-photon regime and tunneling regime depending on the Keldysh \( \gamma \) parameter [3.15], defined as \( \gamma = (E_i/2\Phi)^{1/2} \). Here \( E_i \) is the ionization potential of the atom or ion and \( \Phi \) is the ponderomotive potential of the laser, which is simply the average kinetic energy of an electron oscillating in the electric field of the laser. Given the laser frequency \( \omega_0 \), and its peak amplitude of electrical field \( E_0 \), ponderomotive potential can be written as:

\[
\Phi = \frac{1}{4} m \left( \frac{qE_0}{m\omega_0} \right)^2 = 9.33 \times 10^{-14} I[W/cm^2] \lambda^2 [\mu m]
\]  
(3-1)

Here, \( I \) is the intensity of the laser pulse and \( \lambda \) is the wavelength. The Keldysh parameter can also be written in practical units as:

\[
\gamma = 2.3 \times 10^6 \sqrt{\frac{E_i[eV]}{I[W/cm^2]}} \frac{1}{\lambda[\mu m]}
\]  
(3-2)
Another definition of Keldysh parameter is $\gamma = \omega_0 \tau_t$, where $\tau_t$ is the tunneling time, i.e., the transit time of the electrons tunneling through the atomic Coulomb barrier. This definition draws more physical insights in distinguishing multi-photon and tunneling regimes in OFI ionization.

**Multi-photon regime** ($\gamma > 1$): In this case, the electrons cannot tunnel through the Coulomb barrier within one cycle of the laser oscillation. The ionization, with ionization energy $E_i$, occurs due to the simultaneous absorption of $m$ photons, each of which has an energy $h \nu$, so that the total amount of energy gained from the laser field exceeds the ionization potential ($m h \nu > E_i$). This process can be described theoretically in terms of a matrix element $\langle f | V | i \rangle$, where $|i\rangle$ and $|f\rangle$ are the initial and final states respectively. $V$ is the interaction that couples the electrons with the radiation field and is usually taken as a perturbation. At least $m$-th order terms from the perturbation expansion have to be considered for a correct illustration. The multi-photon ionization rate in s$^{-1}$, or probability of ionization per unit time, is given by [3.16]:

$$W_m = \left( \frac{\sigma I}{h \omega_0^2} \right)^m \left[ \frac{2 \pi \omega_0}{(m-1)!} \right]$$  \hspace{1cm} (3-3)

where $\sigma = 10^{-16}$ cm$^2$. This rate is proportional to the $m$-th power of the laser intensity $I$, provided that resonances with intermediate atomic states are absent. Take carbon atoms (CI, $E_i=11.26$ eV) as an example. For laser pulses of 800 nm ($h \nu = 1.56$ eV), at least 8 photons have to be absorbed ($m=8$) in order to strip one electron from CI. Therefore, the dominance of multi-photon ionization for 100 fs pulses to create plasmas from carbon atoms requires an intensity of $10^{12}$-$10^{13}$ W/cm$^2$, which is easily accessible in current technology, e.g. a 0.1 mJ, 100 fs pulse focused into a 100 $\mu$m spot.
Tunneling regime ($\gamma<1$): Smaller $\gamma$ means stronger laser field, which can suppress the binding field of atoms or ions so that bound electrons can have a considerable magnitude of probability to overcome the bondage by tunneling effects. In its extreme case (usually $I>10^{16}$ W/cm$^2$), the laser field can even totally surpass the Coulomb field and thus permits electrons to escape freely. The ionization rate, calculated by considering an atom or ion in the presence of a static electric field of amplitude $E_0$, is given as [3.16]:

$$W_i = 1.6 \times 10^{17} \left( \frac{E_i}{E_H} \right)^{7/4} \exp \left( -\frac{2}{3} \left( \frac{E_i}{E_H} \right)^{2/3} / E^* \right) \left( E^* \right)^{1/2}$$  \hspace{1cm} (3-4)$$

Here $E^*=E_0/E_a$ is the normalized electric field of the laser pulse, $E_a=q/r_B^2=5.2\times10^9$ V/cm is the Coulomb field in the hydrogen atom with $r_B$ as its Bohr radius, $E_H=13.6$ eV is the ionization potential of hydrogen atom. We can define the tunneling ionization time as $1/W_i$, which characterizes the time scale during which the bound electrons in certain state of atoms or ions are stripped out.

Nitrogen and hydrogen are the two gases used in our investigation of the ultrashort pulse propagation in this chapter. In Figure 3-1, we plot the ionization rate versus the laser intensity for hydrogen atom ($E_i=13.6$ eV) and nitrogen sequence from NI to NV, whose ionization potentials are 14.5 eV, 29.6 eV, 47.4 eV, 77.8 eV and 97.9 eV respectively. We can see that the ionization rate, presented in a logarithmic scale, is very sensitive to the laser intensity for a given ionization potentials and there usually exists a threshold intensity above which efficient ionization occurs. From Figure 3-1, in order to strip one more electron from NV ions within 120 fs (our pulse duration), a minimum intensity of $3-4\times10^{16}$ W/cm$^2$ is needed. The created NVI ions have two electrons left
$E_i=552$ eV and 667 eV respectively) and are in the so-called bottleneck ionization stage. Therefore, it would take a much higher intensity ($>10^{19}$ W/cm$^2$) for further ionization. We are also interested in the ionization carbon, which needs to be totally stripped in order to produce sufficient CVI ions through three-body recombination in recombination soft x-ray lasers. Similar to nitrogen, the first four electrons of carbon can be readily ionized with an intensity of $10^{16}$ W/cm$^2$ while stripping the last two ($E_i=392$ and 490 eV respectively) requires at least $5\times10^{18}$ W/cm$^2$.

![Figure 3-1](image)

**Figure 3-1.** Tunneling ionization time as a function of laser intensity for hydrogen atom (‘H’) and nitrogen atom/ions of different ionization stage (‘N I’ to ‘N V’)

3.1.2 Ionization-induced refraction and scattering instability

The highly nonlinear dependence of ionization rate on laser intensity creates a significant obstacle that prevents the ultrashort pulses from propagating a distance longer than its Rayleigh length in gases and plasmas. Consider a Gaussian pulse in uniform
gases, as the laser intensity sharply decreases from the center toward the fringe, so does the ionization rate. Therefore, the resulted plasma density peaks on axis and diminishes outward. The refractive index experienced by a laser with frequency $\omega_0$ in plasmas of electron density $n_e$ is:

$$n(r) = \sqrt{1 - \frac{\omega_p(r)^2}{\omega_0^2}}$$

(3-5)

Here $\omega_p$ is the plasma frequency depending on electron density:

$$\omega_p = \frac{n_e(r)e^2}{\epsilon_0 m_e}$$

(3-6)

To the opposite of the density distribution, the refractive index has its maximal value at the edge of the plasma and reaches its minimum on axis. Because ionization usually occurs at the leading edge, the rest of the pulse is in fact propagating in the hitherto created plasma, instead of neutral gases. Smaller on-axis refractive index allows the central part of the pulse to travel faster than the fringe, distorting the laser wave front as shown in Figure 3-2. The net effect is that plasmas behave like a negative lens which refracts the pulses away from the axis, initiating the ionization induced refraction.

![Figure 3-2](image), Illustration of ionization induced refraction; the plasma column (red) has maximal density on axis. Solid line indicates the wave front while dashed arrows show the beam propagating direction
We can define a refraction length $L_{\text{ref}}$ after which the central part of the pulse outruns the fringe by a phase of $\pi/2$ [3.17]:

$$L_{\text{ref}} = \frac{1}{2} \frac{\lambda_0}{n_c} \frac{n_c}{n_e}$$  \hspace{1cm} (3-7)

Where $n_c$ is the critical density:

$$n_c = \frac{\mathcal{E}_0 m_e \omega_0^2}{e^2} = 1.12 \times 10^{21}[\text{cm}^{-3}] \sqrt{\frac{\mathcal{E}}{\mu \text{m}}^2}$$  \hspace{1cm} (3-8)

Equating $L_{\text{ref}}$ to the Rayleigh length of the pulse $z_R = \pi r_0^2/\lambda$, which physically means the refraction effect is comparable to the natural diffraction of the pulse, sets a limit on the maximal achievable electron density $n_e$, given by:

$$\frac{n_e}{n_c} = \frac{\lambda_0^2}{2\pi r_0^2}$$  \hspace{1cm} (3-9)

Similar to the plasma density, the pulse intensity cannot go beyond a clamping value either. The ionization-induced refraction starts refracting the pulse out as long as its intensity exceeds the ionization threshold and prevents the pulse from reaching the same focus, or peak intensity as in vacuum. This can also be interpreted by examining the focal spot size of the laser pulse through the combination of focusing lens and plasmas. Plasmas, as a negative lens, increase the $f$ number ($F/#$) and thus the focal spot size, practically reducing the peak intensity the pulse can reach. All these effects prohibit the laser pulse from reaching and maintaining a high intensity over a long distance and thus limit the length of the plasma that can be created by ultrashort pulses.

The nonlinear ionization rate can also trigger the ionization scattering instability [3.18], in which the modulations of the electron density transverse to the pulse propagating
direction are collectively amplified. The inhomogeneous ionization rate might create a plasma density modulation in radial direction that scatters the laser pulse. The scattered radiation in turn reinforces the modulation of the laser field, leading to a positive feedback loop, \emph{i.e.} instability. This instability can grow very fast and disrupt the propagation of laser pulse within a short range of time and distance. The ionization-induced refraction worsens this situation since the pulse will become more unstable to the scattering instability after being refracted [3.18]. However, under certain proper conditions, the refraction can be compensated either by self-channeling (Kerr effect, relativistic self-channeling, ponderomotive self-focusing, \emph{etc}) or with the help of a pre-formed plasma waveguide. They are discussed in detail in Chapter 4.

3.1.3 Raman Forward Scattering (RFS) Instability

Unlike ionization scattering instability which starts from the transverse modulation of plasma density, RFS instability is seeded by the density modulation in longitudinal direction. RFS is a resonant decay of a strong electromagnetic wave, with frequency and wave number \((\omega_0, k_0)\), into a scattered wave \((\omega_s, k_s)\) and a plasma wave \((\omega_p, k_p)\), both of which are propagating in the same direction (as its name “forward” indicates) as the original wave. Energy and momentum conservation has to be satisfied as:

\[
\omega_0 = \omega_s + \omega_p \\
kd = kd + kp
\]  

(3-10)

Considering that any radiation traveling inside the plasmas must have a frequency larger than \(\omega_p\), the minimum frequency of the incoming pulse has to be \(\omega_0 \geq 2\omega_p\), \emph{i.e.} RFS only occur in plasmas whose density is lower than a quarter of the critical density \(n_e \leq 1/4n_c\).
The onset of RFS instability can be interpreted as the mutual reinforcement between the scattered wave and the longitudinal density modulation [3.19]. Consider a pulse with field amplitude $E_0$ propagating in plasmas with longitudinal density modulation $\delta n$ associated with plasma wave. The transverse motion of electrons in the laser field generates a current density $\delta J = -eV_L\delta n$ where $V_L = -eE_0/(m\omega_0)$, and a scattered wave with field amplitude $E_s$. The spatial and temporal interference of the scattered wave with the incoming laser beam produces an electromagnetic beat envelope of frequency $\omega_p$, propagating synchronously with the plasma wave. This beat wave reinforces the original density modulation through ponderomotive force that is proportional to $\nabla (E_0 \cdot E_s)$. The enhanced $\delta n$ then scatters more incident light into Raman bands ($\omega_s$, $k_s$) and initiate instability through this feedback loop.

RFS instability may grow from density fluctuation created by thermal noise, or nonlinear force exerted by the laser pulse itself. For ultrashort pulses, the major source of seeding is the wake excited by the ponderomotive force of an ionizing front in the leading edge of the laser pulse. This mechanism was first discussed by Mori et al [3.20] and then verified in detailed simulation [3.21] and experiments [3.22]-[3.23].

The spatiotemporal gain of the RFS instability for arbitrary laser beam intensity can be written as [3.23]:

$$G = \exp(g) \left( \frac{2}{\pi g} \right)^{1/2}$$

(3-11)

Here:

$$g = \left[ \frac{a_0}{\sqrt{2(1 + a_0^2/2)}} \right] \left( \frac{\omega_p}{\omega_0} \right)^2 \left( \frac{\omega_0}{c} \right) \sqrt{x\varphi}$$

(3-12)
where $a_0$ is the normalized vector potential of the laser pulse defined as:

$$a_0 = eA/mc^2 = 0.86 \times 10^{-9} \lambda \mu m \sqrt{I_0[W/cm^2]}$$

(3-13)

$\omega_0$ and $\omega_p$ are the frequencies of the laser and plasma waves respectively, $x$ is the distance traveled in the plasma, and $\varphi/c$ is the length of time that the assumed constant-intensity pulse has interacted with the plasma at positive $x$. The $(1+a_0^2/2)$ factor is the relativistic correction for ultrahigh intensity pulses. In extremely strong laser field ($a_0 \geq 1$), the oscillating motion of electrons becomes relativistic, which means the electron mass is dependent on the oscillating speed determined by the laser intensity. Accordingly, plasma frequency, a function of electron mass, also relies on the laser intensity and a correction factor has to be introduced to describe this effect.

Figure 3-3, Gain of RFS instability versus laser intensity for three different plasma densities related to our experiment, laser pulse is 120 fs long, 1.5 mm Rayleigh length
We plot the RFS instability gain (the total amplification) as a function of laser intensity in Figure 3-3. The values of $x$ and $\varphi/c$ are 1.5 mm and 120 fs, corresponding to the Rayleigh length and pulse duration in our experiment, respectively. Three different plasma densities relevant to our experiment are being used and it is clear to see that the instability is quite sensitive to the plasma density. Notice that for the intensity range and wavelength ($\lambda=800$ nm) considered here, the maximal $a_0$ is $\sim0.22$ so the laser is not yet entering the strongly relativistic regime. Nevertheless, the calculated gain, as the intensity approaching $10^{17}$ W/cm$^2$ that is used in the experiment discussed in this chapter, is definitely not negligible and RFS instability is very likely to disturb the propagation of the 120 fs pulse. Both ionization scattering and Raman forward instability can be effectively suppressed using a pre-formed plasma waveguide. The density barrier of the waveguide can confine the laser pulse within its radius, therefore terminating the positive feedback loop that leads to ionization scattering instabilities. As for Raman forward instability, the growth rate is reduced and the scaling with the laser intensity is modified in plasma waveguides because of the radial shear of the plasma frequency and the existence of weakly damped hybrid (electrostatic / electromagnetic) modes of the radially inhomogeneous plasma [3.24]. This method is discussed in detail in Chapter 4.

3.2 Experimental Setup

3.2.1 Overview

The Ti:Sapphire laser system Thales Alpha with output pulses of 1.4 J in 120 fs, at 800 nm and a repetition rate of 5 Hz has been used to create plasmas in the experiments,
while the probe beam for the diagnostics of neutral gas and plasma density was from the Positive Light by frequency doubling its output at 1064 nm to 532 nm with a pulse duration of 200 fs. For studying the dynamics of plasma formation in gases, we limited the laser output to 100 mJ in 120 fs in order to avoid self-phase modulation in optical elements in the current setup. The laser beam was focused into a spot size of about 40 $\mu$m with a spherical lens of $f=30$ cm (F/15), which gave a Rayleigh length of 1.5 mm. Peak intensity close to $4\times10^{16}$ W/cm$^2$ is estimated from the above parameters.

![Experimental setup for the investigation on propagating a 120 fs pulse through a gas jet](image)

**Figure 3-4.** *Experimental setup for the investigation on propagating a 120 fs pulse through a gas jet*

The plasma was created in a supersonic gas jet using a pulsed gas valve developed at the Institute of Optoelectronics, Warsaw [3.25] and its schematic is shown in Figure 3-5. The valve has a reservoir backed with gases under the pressures up to 400 psi. The reservoir was closed with a diaphragm driven by an electromagnetic coil. Usually we ran the valve at a repetition rate no higher than 0.5 Hz (one shot every two seconds) in order
to avoid the melting of fuses in the power supply which delivered high current pulses to the coil. We have used a rectangular orifice of $6 \text{ mm} \times 0.5 \text{ mm}$, which allowed us to change the effective length of the gas target by changing the orientation angle of the orifice with respect to the laser propagation axis. The gas density was controlled by changing the backing pressure of the valve or its opening timing. Here we delivered the laser pulse across the orifice of the gas jet in order for simultaneous monitoring of both neutral gas and plasma densities during each laser shot.

![Figure 3-5](image)

**Figure 3-5, Schematic of an electromagnetic valve to create gas puff targets**

The major diagnostic tool in this experiment was a shear-type interferometry, which utilized the dependence of refractive index of probe beam on particle densities. The methods to generate interference may vary (Michelson type, Mach-Zehnder, shear type, etc), but the principles under which particle densities are extracted from the interferogram are basically the same. In this experiment, the interference pattern was produced by overlapping two parts of a single collimated probe beam. One part of the beam passed through the plasma region (signal beam) whiles the other part (reference beam) carrying the reference phase bypassed the gas jet. Both parts were combined on CCD detector...
using bi-prism and collimating lenses. The difference in optical paths of the two parts introduced a phase difference, which was visualized as certain bending on the otherwise straight interference fringes if no gas or plasma was present. Section 3.2.2 describes in detail how the density information can be acquired from the bending of fringes.

An interferometric 400nm band pass filter was placed in front of the CCD to prevent plasma emission from reaching the detector. Since both parts of the probe beam passed through the same optical elements, the whole system was quite robust against mechanical or acoustic noise, which made possible absolute phase shift measurements on a shot-to-shot basis. The time resolution was limited by the probe pulse duration which was 200 fs while the spatial resolution was about 5 $\mu$m. The synchronization and adjustment of delays between the laser pulse and the probe have been controlled with micrometric precision by changing the optical delay line in the probe pass. Good reproducibility of the measurements allowed snapshots of the plasma interferograms taken at different probe delays to be combined into a movie, thereby displaying the propagation of the laser followed by the plasma channel expansion.

3.2.2 Measurement of neutral gas and plasma density

In our experiment, the neutral gas and plasma densities are measured by the same interferometer and its working principle is described below.

**Gas density measurement:** Consider here a probe beam at a wavelength of $\lambda$ passing through a gas jet with density $N$ and length $L$. The refractive index of the probe beam can be written as: $n(N)=1+\Delta n=1+\alpha N$, where $\alpha$ is a coefficient depending only on gas species. At 1 atm, we can usually find published data of the refractive index $n_0=1+\Delta n_0$ and thus
derive $\alpha = \Delta n_0 / N_0$, where $N_0=N_a/22.4L=2.69 \times 10^{19} \text{ cm}^{-3}$ is the gas density also at 1 atm in room temperature and $N_a$ is the Avogadro constant. After passing the gas jet, the two parts of the beam have a phase difference $\Delta \varphi = (2\pi / \lambda)(n - 1)L = (2\pi / \lambda)\alpha NL$, causing the corresponding fringe on the interferogram to shift by an amount of $\Delta N_f$ (phase shift is then $\Delta \varphi = 2\pi \Delta N_f$). Here we assume that gas density along the beam path is uniform since the jet length (6mm) is much longer than the characteristic length of the gas density gradient at the edge (<0.5 mm). Equating the two expression of phase shift $\Delta \varphi$, we can write the gas density as:

$$N = 2.69 \times 10^{19} \frac{\Delta N_f \lambda}{L \Delta n_0} \text{ cm}^{-3} \quad (3-14)$$

For example, for $L=6 \text{ mm}$, $\lambda=400 \text{ nm}$, $\Delta n_0$ of nitrogen is $2.98 \times 10^{-4}$, then 1 fringe shift ($\Delta N_f=1$) corresponds to a gas density of $6 \times 10^{18} \text{ cm}^{-3}$.

In practice, we number the fringes with $N_i$, $i=1, 2 \ldots i_0$, on two interferograms, where $i_0$ is the total number of fringes on each image. One interferogram is with the jet on (information image) and the other is taken when the jet is turned off (reference image, fringes are straight lines). The acquired $p_i(N_i)$ relation gives the pixel position $p_i$ for each fringe $N_i$. Then through interpolation, we derive the $N_j(p_j)$ mapping that tells us the fringe number for each pixel (notice that $N_j$ is not necessarily an integer since each fringe consists of multiple pixels, usually 10-30). Finally we subtract the two mappings, achieving the phase-difference and thus the density distribution.

**Plasma density measurement:** The refractive index of plasmas depends on the plasma density as:

$$n = \left(1 - \frac{\omega_p^2}{\omega_0^2}\right)^{1/2} = \left(1 - \frac{n_e}{n_c}\right)^{1/2} \quad (3-15)$$
Therefore, the phase difference between the two parts of the interferometric beam is:

\[ \Delta \varphi = \frac{2\pi}{\lambda} \int_{r_1}^{r_2} (n-1)\,dl = \frac{2\pi}{\lambda} \int_{r_1}^{r_2} \left[ \left(1 - \frac{n_e}{n_c}\right)^{1/2} - 1 \right] \,dl \]  

(3-16)

Here \( r_1 \) and \( r_2 \) are the entrance and exit points of the interferometric beam in the plasma and the line integral is along the beam. For underdense plasmas, where \( n_e \ll n_c \), we can approximate the refractive index to the first order \( \frac{1}{2} n_c n - \frac{n_e}{n_c} \) and thus the phase difference can be simplified as:

\[ \Delta \varphi = \frac{\pi}{\lambda n_c} \int_{r_1}^{r_2} n_e \,dl \]  

(3-17)

We can define an average density \( \tilde{n}_e \), which is constant along the beam path and introduces the same amount of phase shift \( \Delta \varphi \). In this case the integral is simply the product of the average density and plasma diameter \( D \), and \( \tilde{n}_e \) can be written as:

\[ \tilde{n}_e = \frac{n_e \Delta \varphi}{\pi D} \]  

(3-18)

Therefore, the procedure of acquiring the phase and average density of plasmas is the same as of gases. The plasmas diameter \( D \) can also be found from interferograms where the boundary of the plasma is clearly defined by the front of the expanding shock waves.
In some cases, particularly those involving the propagation and guiding of powerful pulses in underdense plasmas, the radial distribution of plasma density is critical. Unfortunately, for interferometers, the acquired phase shift is always integrated over certain lengths so the plasma density at certain specific points is not directly measurable.

However, if we assume a radially symmetric density, i.e., $n_e = n_e(r)$, which is a quite good approximation in our experiment, then the radial density distribution can be recovered by Abel transformation as illustrated in Figure 3-6. Consider a probe beam path with a distance $y$ above the center of the plasma. For each point on this line, the density is only dependent on $r$, the length to origin. So the total phase shift $\Delta \phi$ over this line, and the recovered density at each point are:

$$\Delta \phi(y) = 2 \int_y^{R_p} n_e(r) \frac{r}{\sqrt{r^2 - y^2}} dr$$  \hspace{1cm} (3-19)

$$n_e(r) = -\frac{1}{\pi} \int_r^{R_p} d\Delta\phi(y) \frac{dy}{\sqrt{r^2 - y^2}}$$  \hspace{1cm} (3-20)
In our experiment, the probe beam is much larger than the plasma cross section, so a single shot of the probe beam can give us a phase-difference distribution $\Delta \phi(y)$ over all possible $y$ values. In case that the probe beam cannot cover the whole plasma diameter, multiple beams or a spatial scanning across the plasma will provide sufficient information on the density distribution.

3.3 Experimental Results and Discussion

3.3.1 Characterization of the gas jet

As described in the introduction of this thesis, a variety of targets including solid materials, micro-capillaries, gas cell and gas jet have been used in soft x-ray laser experiments. Comparing with solid target, gas jet is free of any ablation debris that might pose serious potential damages to optics; it also provides fresh plasma for each shot, thus avoid the problems associated with crust formation over the solid surface; moreover, plasma parameters in gas jets can be well monitored so we can keep track of the experimental status. The advantage of gas jet over static gas cell is its limited spatial extent which effectively alleviates, though not totally eliminates, the refraction, self-focusing and various other nonlinear process that would disrupt the propagation of high-intensity laser pulses in the target.

A sample measurement of neutral particle densities is illustrated in Figure 3-7 for the case of the nitrogen jet. The original interferogram can be used to obtain a phase shift map by subtracting the reference interferogram taken when the jet valve was closed. The dark circles in Figure 3-7 (b) are equal-density contours, with each circle marking one
more fringe shift toward the center. For example, the white dot in the graph is between the 4th and 5th circle, so we can estimate the gas density at that point is between 2.4-3×10^{19} \text{ cm}^{-3} according to formula (3-14).

Figure 3-7, Interferometry at 400 nm in N\textsubscript{2} jet: (a) the interferogram of jet around 50 µs after valve opening; (b) after subtraction of reference interferogram taken with closed valve (phase shift map)

Figure 3-8, Gas (C\textsubscript{2}H\textsubscript{6}) density distribution across the nozzle for different jet opening time, probe beam is 0.5 mm above nozzle
More detailed characterization is calculated from local fringe shifts using available data on optical densities of gases [3.26]. Errors associated with the uncertainly in the gas length L (~6mm) are expected to be negligible in the region of interest (i.e., within a perimeter of 0.5 to 1 mm outside the perimeter of the orifice). The dependence of gas density on two parameters—jet opening time and distance from the nozzle surface—is of our particular interest. By changing the jet opening time, we are actually changing the gas density at the moment when the ionization pulse arrives, i.e., the ionization pulse will “see” different gas densities for different jet opening time. The distance from nozzle surface plays a slightly more complex effect on the gas density. When moving away from the surface of the nozzle, the spreading of the gas not only reduces the density, but also elongates the laser-gas interaction region, which is characterized by the FWHM (Full Width of Half Maximum) of the density distribution.

![Figure 3-9](image_url), Peak density of C$_2$H$_6$ jet at 0.5 mm above the surface of the nozzle as a function of jet opening time.
Figure 3-8 shows the density distribution of the \( \text{C}_2\text{H}_6 \) gas at 0.5 mm above the surface of the nozzle for different jet opening times, and \( z \) represents the position across the jet (see Figure 3-7). All the distributions have a near Gaussian shape, and the decay of their peak density is illustrated in Figure 3-9. The valve is capable of ejecting gases up to \( 4 \times 10^{19} \text{ cm}^{-3} \), therefore, if fully ionized, providing sufficient ion density for soft x-ray lasers, which typically require \( 10^{18}-10^{20} \text{ cm}^{-3} \). It is worth to mention that although the amplitude of the density distributions drops as the jet opening time increases, a roughly constant FWHM (~0.8 mm) of the distribution is maintained, \( i.e. \) the effective gas length is not affected by the jet opening time. Here we are only presenting the data within a time span of 0.25 ms, shorter than the duration of the jet opening that is approximately 1 ms, since our primary concern lies in the effective adjustment of the gas density, instead of the long plateau region before 200.15 ms as can be seen from Figure 3-9.

**Figure 3-10, Gas \( \text{C}_2\text{H}_6 \) density distribution for different distances from the surface of the nozzle, jet opening time is fixed at 200.15 ms**
Figure 3-11. Peak density of C$_2$H$_6$ jet as a function of distance above nozzle, jet opening time is fixed at 200.15 ms

In Figure 3-10, we plot the density distribution of C$_2$H$_6$ jet at difference distances from the surface of the nozzle, with a fixed jet opening time at 200.15 ms. A key feature of the density evolution is the spreading of the spatial extent. At 0.31 mm above nozzle, the FWHM of the gas puff is around 0.5 mm, and this value increases up to more than 1 mm when the measurement point is 1.5 mm away from the nozzle surface. The aftermath of the spatial-extent elongation is twofold: it increases the interaction length for the ionization pulse, but at the same time introduces or aggravates a variety of nonlinear processes before the pulse reaches the center of the jet. The vacuum-gas interface is no longer sharp at a distance 1 mm or further away from the nozzle, so ionization occurs before the pulse reaches the high density area and imposes detrimental effect on the pulse propagation in general. Figure 3-11 shows us the peak densities as a function of distance from nozzle surface. The decay rate is noticeably faster than that of linear decay. Within
the parameter window considered here, the density decay is roughly following an exponential form $N(d) = ad^{-b}$ where $d$ is the distance from the surface of the nozzle, $a$ and $b$ are constant coefficients and $b$ is close to unity.

3.3.2 Pulse propagation in gas jet

Three different gases have been used in the experiments: nitrogen ($\text{N}_2$), ethane ($\text{C}_2\text{H}_6$), and hydrogen ($\text{H}_2$). This chapter focuses mainly on the data for $\text{H}_2$ and $\text{N}_2$ jets, and trying to ascertain the physics underlying the distinctive propagation behaviors between these two jets. The primary features for $\text{C}_2\text{H}_6$ data are similar to $\text{N}_2$ so it is not presented here.

![Figure 3-12](image_url)

**Figure 3-12.** Evolution of plasma created in (a) nitrogen and (b) hydrogen gas jets by 90mJ, 120fs laser pulse. Initial molecular density in jets is represented by color scale. The rectangular bars indicate the size and location of laser beams and the arrow on the left indicates the direction of the incoming pulse.
Figure 3-13, Evolution of OFI plasma in N$_2$ jet presented by snapshots of interferograms: from top to bottom at 2.33ps, 3.0ps, 3.33ps, 4.33ps, and 5.0ps delays indicated by white rectangular bars presenting the position of the laser pulse. Electron density irregularities are marked with arrows, while the block arrow on the left indicates the direction of the incoming pulse; (b) The distribution of initial nitrogen molecular density along the propagation axis.

The interferograms shown in Figure 3-12 for N$_2$ and H$_2$ jets are presented at different delays of the probe pulse. The rectangular reference bar on each interferogram indicates the location of the driving laser pulse. Its dimensions on the pictures approximately correspond to the length and diameter of the laser pulse in space. The position of the laser pulse is established by the optical delay measurement of the probe pulse, which is controlled by the delay line. The origin of time (t=0) is chosen as the moment at which the plasma just appears on the interferogram. The OFI process suggests ionization takes
place within the instantaneous location of the laser pulse. However, to avoid obstruction of view and for convenience the reference bar is shown slightly shifted ahead of the leading edge of the plasma. Apparently, extension of the plasma follows almost exactly the propagation of the pulse, since the position of the bar remains steadily near the leading edge of the plasma until the moment when the plasma channel stops to increase in length (this phenomenon is discussed in detail below).

**Figure 3-14.** Evolution of average electron density in (a) \(N_2\) and (b) \(H_2\) gas jets. The maximum expected value of plasma density (dashed lines) along the laser propagation axis calculated from measured initial gas density in assumption of getting 10 electrons from each \(N_2\) molecule and for totally stripped of hydrogen atoms. The actual delay for each density profile is represented by position of roof mirror in the optical delay line.

Comparing the interferograms for \(N_2\) and \(H_2\) jets, one can see the distinct difference between these two cases: the plasma is noticeably wider and shorter in \(N_2\) than in \(H_2\). In
the case of H₂ it is formed as a long channel with a comparable diameter to the laser beam. Furthermore, in the N₂ plasma the ionization front at the leading edge of the pulse exhibits irregularities—spikes and dents over smooth background fringes—which are especially pronounced when the jet is operating with higher gas densities. That case is illustrated in Figure 3-13 where some irregularities are indicated with arrows. This feature of plasma structure is irreproducible from shot-to-shot, which can also be seen on the snapshots presented in Figure 3-13.

In the case of N₂, the plasma is formed mostly near the entrance of the gas jet regardless of the initial gas density, whereas in H₂ the plasma is created all the way to the opposite side of the jet. This effect is clearly seen in Figure 3-14, where the average plasma density evolution along the axis is presented for nitrogen and hydrogen. The expected electron densities calculated from the measured neutral gas density have been plotted there for comparison. The dashed curve in Figure 3-14 (a) represents the plasma density assuming 5 electrons are stripped from each nitrogen atom (10 electrons from each N₂ molecule). Near the beam entrance the measured electron density agrees well with the expected stage of ionization of nitrogen given the high ionization potential of the two remaining 1-s electrons in N⁵⁺ ions. In the case of H₂, as expected, the plasma is fully ionized (Figure 3-14 (b)). The achieved maximum plasma densities for both hydrogen and nitrogen are close to 10²⁰ cm⁻³ (Figure 3-14). Therefore, naturally, one expects no significant difference in refraction between the two cases. Yet, in the case of N₂, refraction is apparently more significant. Moreover, in experiments even with reduced initial N₂ density, where the electron density is lower than that of H₂ case, the propagating distance of the laser beam is still shorter.
It seems that the major difference in laser beam propagation in hydrogen and nitrogen plasmas is related to the ionization energy of the total number of atoms of H and N along the path of laser beams. Neglecting the dissociation potential of molecules, in hydrogen it is 13.6 eV, whereas for nitrogen, the total energy required to release the first 5 electrons is \( \sim 267 \text{eV} \) (14.5+29.6+47.4+77.7+97.9 eV). The ionization potential for the remaining two electrons (552 eV, and 667 eV, respectively) is too high for tunneling ionization to be significant at \( 4 \times 10^{16} \text{ W/cm}^2 \). In the time domain, different stages of ionization are realized through the mechanism of tunneling ionization at different times during the laser pulse: when the optical field becomes comparable to the binding field of an electron in an atom or ion, that electron is released. In fact, when the external field reaches 10% of the binding field, it takes about 3 optical cycles to ionize the atom/ion [3.27]. The ionization thresholds for the first 3 electrons of nitrogen atoms are reached well before the laser pulse maximum. The 4th and 5th electrons are released from N\(^{3+}\) and N\(^{4+}\) ions at the moment just before the maximum of the laser pulse.

Similar considerations apply in the space domain. The first 3 electrons are released in an area larger than the laser beam diameter, while the 4th and 5th electrons from ionization of N\(^{3+}\) and N\(^{4+}\) are released near/within the waist of the laser beam, as can be indicated from the ionization rate plot in Figure 3-1. In the case of hydrogen, ionization is limited entirely to the region inside the beam diameter, and therefore most of the beam propagates in relatively uniform plasma and does not experience significant refraction. In contrast, for nitrogen, the 4th and 5th electrons are released just before the pulse maximum, and the core of the pulse propagates in a region having two steps in plasma density related to two stages of ionization. The whole density profile may be
significant altered due to refraction of the beam, and consequently the propagation length of the laser beam is affected.

3.4 Results of Computational Modeling

The significant difference in plasma length in H₂ and N₂ jets suggests that it is related to the effect of the gas ionization during pulse propagation. In order to better understand this phenomenon, we have closely collaborated with Dr. Phillip Sprangle and Dr. Dan Gordon from the Plasma Division at Naval Research Laboratory (NRL) and carried out several large scale, three dimensional particle-in-cell (PIC) simulations using turboWAVE [3.28]. Some of the simulations utilized the ponderomotive guiding center (PGC) technique, which makes approximations based on the disparity between the laser and plasma frequencies [3.28]. The rest of the simulations used fully explicit PIC, which describes the laser pulse using the exact Maxwell equations. Of these, some neglected the absorption of laser radiation due to direct ionization losses, while others included this effect. In all cases, the ions and neutral atoms were held immobile and molecular processes were neglected. The computational region was set in motion at the speed of light in order to follow the laser pulse. Perfectly matched layers were used to absorb radiation incident on the transverse boundaries. Ionization was modeled using a Monte-Carlo approach, with the ionization rate being derived from the Ammosov-Delone-Krainov (ADK) [3.29] formula. Ionization effects in laser plasma interactions have also been discussed in [3.30]-[3.33].

The parameters common to all the simulations reported here are as follows. Propagation is in the z-direction, while polarization is in the x-direction. The gas density
vanishes for $z < 0$, rises smoothly to its peak value at $z = 0.75$ mm, and falls back to zero again at $z = 1.5$ mm (see, e.g., Figure 3-18). The gas density is independent of $x$ and $y$. The temporal shape of the laser amplitude is the same as the gas density profile. The intensity full-width at half-maximum (FWHM) is 100 fs. The laser pulse is assumed to be Gaussian, with a spot size radius ($1/e$ of the amplitude) of 40 $\mu$m (this corresponds to an intensity distribution with FWHM of 47 $\mu$m). The peak intensity of the laser pulse was $4 \times 10^{16}$ W/cm$^2$. The simulations are initialized with the peak of the laser pulse at $z = -39$ $\mu$m.

![Figure 3-15](image)

**Figure 3-15.** Comparison of (a) PGC simulation and (b) fully explicit simulation of pulse propagation in tunnel-ionized nitrogen. The data is evaluated at $t = 1$ ps, before any severe disruptions take place. $E_{br} = mc\omega_p/e$ is the cold wave breaking field.

The PGC technique was found to be useful only during the early stages of the pulse evolution. Late in time, scattering of radiation into large angles, as well as the development of large nonlinear frequency shifts, begins to violate the assumptions of the
PGC model. Comparison of the results from the two models at earlier time is shown in Figure 3-15 for the case of the N\textsubscript{2} jet. The parameters of the simulations were the same as in Figure 3-14. After 0.35 mm of propagation, a staircase intensity pattern develops due to ionization induced steepening in the presence of multiple ionization fronts [3.18]-[3.21]. The location of the ionization fronts is sometimes more obvious in the fully explicit data. In fact, ionization induced refraction [3.32] can be clearly seen by comparing the phase fronts in the core with the phase fronts just outside the core. Considering that the grey scale is logarithmic, it is clear that only the fifth ionization front refracts significant pulse energy. Similar runs were done in hydrogen, where it was seen that ionization-induced distortions only occur in the radial or temporal extremities of the pulse, \textit{i.e.} the edge or the end of the pulse.

Figure 3-16, Laser electric field after propagation in nitrogen for (a) 1.8 ps and (b) 3.3 ps. $E_{br} = m\omega_p/e$ is the cold wave breaking field. The back third of the computational region is not shown for illustrating purposes.
Figure 3-17, Electron density due to tunnel ionization of nitrogen atoms after (a) 1.8 ps propagation and (b) 3.3 ps propagation. The thin vacuum regions near the edges of the computational box are due to the perfectly matched layers. In reality the plasma would extend beyond the transverse boundaries of the box. The back third of the computational region is not shown for illustrating purposes.

At longer propagation distances, the pulse develops in a complicated way, due to at least two additional processes. First, the presence of axial ionization fronts accelerates the growth of forward Raman instabilities [3.21]. This is due to the ponderomotive force associated with the steepened laser pulse profile [3.21], and the effective force that arises from the dynamics of tunnel-ionized electrons [3.20]. Second, the presence of radial ionization fronts leads to the ionization scattering instability [3.18]. The ionization scattering instability leads to the growth of modes with large transverse wave number. The pulse distortions caused by these instabilities are illustrated in Figure 3-16. Figure 3-16(a) shows an image of the laser electric field at $z = 0.54$ mm. The modulations in the core of the pulse have a wavelength consistent with forward Raman scattering.
horizontal striations in the wings are evidence of the ionization scattering instability. Figure 3-16 (b) shows the laser electric field at $z = 0.99 \, \text{mm}$. The pulse consists of filamentary structures, which are probably due to a complex interplay between forward Raman scattering and the ionization scattering instability.

![Graph of electron density](image)

**Figure 3-18.** Axial variation of electron density in nitrogen jet. The maximum expected value (based on quintuple ionization of nitrogen atoms) is shown as a dashed line

The electron densities at the same two timing points as in Figure 3-16 are shown in Figure 3-17. The plasma wave associated with forward Raman scattering can be seen in Figure 3-17 (a), and the filamentary plasma associated with the ionization scattering instability can be seen in Figure 3-17 (b). The predicted filamentary structure in plasma is confirmed also by experiments where irregularities in fringes are observed (Figure 3-13). Due to small size of the filaments their contributions to the measured fringe shift is relatively small. However, their observations are somewhat obscured by limiting spatial
resolution (~5µm) of measurements. Note also that the transverse dimensions of the plasma extend beyond the computational region. Simulations of the hydrogen jet gave the diameter of the plasma of ~115 µm, which is in reasonable agreement with experimental data (Figure 3-12).

From a geometric optics point of view, the rays in the pulse are scattered into large angles in ionization scattering instability, diminishing the intensity of the light as it propagates. As the intensity of the light is reduced, the generation of tunnel-ionized electrons is slowed. This is illustrated in Figure 3-18, which shows the electron density left behind the laser pulse as it propagates through the N₂ jet. For the first 0.5 mm, quintuple ionization occurs. Beyond z = 0.5 mm the density abruptly drops to a value consistent with triple ionization. The run associated with Figure 3-18 accounted for laser absorption due to direct ionization losses. In another set of runs, it was found that suppressing these losses only increases the electron density by about 10%. Simulations were also carried out in a hydrogen jet with peak atomic density \(7 \times 10^{19} \text{ cm}^{-3}\), and in a pre-ionized plasma with peak electron density \(2 \times 10^{20} \text{ cm}^{-3}\). The hydrogen jet was fully ionized throughout, and in both cases there was very little pulse distortion. Thus, ionization enhanced Raman and ionization scattering instabilities appear to be responsible for the drop in plasma density at \(z = 0.5 \text{ mm}\).

Although the fully explicit version of turboWAVE, in principle, can handle arbitrarily large scattering angles, there is a practical limitation due to finite transverse resolution. With present supercomputing technology, it is just barely possible to resolve all the wavenumbers that are generated in the simulation. Poorly resolved modes have artificially slow group velocities. It therefore takes longer for these modes to carry
energy out of the focal volume. This may be one reason the simulation predicts longer plasmas than were seen in the experiment. Another reason might be that the simulation uses a perfect Gaussian laser pulse. Probably the experimental pulse contains higher order modes, which might seed instabilities more strongly.

### 3.5 Conclusions

We have investigated experimentally and theoretically the propagation of sub-picosecond laser pulses in optically ionized hydrogen and nitrogen gases at the intensity of $4 \times 10^{16} \text{ W/cm}^2$ [3.34]. The propagation distance of the laser beam in nitrogen was considerably shorter than in hydrogen. This is due to the strong refraction effect resulting from the stripping of electrons from different ionization stages of nitrogen (NI up to NV) near the laser beam core. Additional mechanisms are the forward Raman and ionization scattering instabilities. The proposed mechanism suggests that the effect of refraction becomes stronger when the laser intensity approaches the threshold of the ATI (Above Threshold Ionization). Therefore, in creating conditions for uniform plasma channels, the reduction of propagation length due to ionization induced refraction has to be taken into account. Such conditions are important for recombination lasers operating on the 2-1 transition of H-like ions in the transient regime. The refraction effect is expected to be significant when the intensity of the pumping laser becomes high enough to ionize the last electrons from the 1s shell. In this case the laser core would propagate in the region with density steps created by the leading edge of the laser pulse and will be subject to strong refraction. However, for the case of H-like carbon at the required laser intensity near $10^{19} \text{ W/cm}^2$, the relativistic self focusing mechanism could become dominant,
favoring creation of an elongated plasma channel. A more promising tool is the plasma waveguide with a hollow-pipe density structure that can effectively guides the propagation of high intensity laser pulses. Chapter 4 discusses in detail the formation of these plasma waveguides.
Chapter 3. Propagation of Ultrashort Pulses in Gas Jet

References:


Chapter 3. Propagation of Ultrashort Pulses in Gas Jet


Chapter 4

Formation of Plasma Waveguides

Propagation of intense laser pulses in gases or plasmas over many Rayleigh lengths has a wide application in contemporary research. Other than soft x-ray lasers discussed in this thesis, it is also closely related to laser-driven charged particle acceleration [4.1]-[4.2], high harmonic generation [4.3]-[4.5], fast ignition scheme of inertial fusion [4.6], and Raman amplification [4.7]-[4.9]. However, as illustrated in last chapter, a variety of nonlinear processes and instabilities, together with the natural diffraction, set severe limitation on the distance the intense pulse can propagate, usually less than the Rayleigh length. Over the past years, much progress has been made in extending the propagation mainly using two methods: self-channeling [4.10] and pre-formed plasma waveguide [4.11]. In this chapter, we first briefly discuss several mechanisms that might lead to the self-channeling of intense pulses, including their advantages and drawbacks. Then we focus on the method of pre-formed plasma waveguides, which can provide stable channeling and superior control over the mode quality of the guided pulses. Experimental results on the formation of plasma waveguides using Bessel and Gaussian beams are presented.
4.1 Overview of Pulse Guiding in Gases and Plasmas

The evolution of laser pulse in ionizing gases or plasmas can be described by the general wave equation:

\[
\left(\nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2}\right) E = \frac{\omega^2}{c^2} \left(1 - n_r^2\right) E
\]

(4-1)

Here, \( E = E(r, z, t) \) is the transverse electric field of the laser pulse and it is a function of both space and time, \( z \) is the propagating direction. \( n_r = n_r(r, z, t) \) is the effective index of refraction, determined by the dynamic response of the medium (gases or plasmas) to the electric field \( E(r, z, t) \) at the temporal and spatial point \( (r, z, t) \). The wave equation indicates that the pulse evolution is solely determined by \( n_r \), the source term in the differential equation. Since numerical techniques are usually required to fully solve equation (4-1) when \( n_r \) is nontrivial, some heuristic analysis becomes very helpful in understanding the key element in pulse guiding.

Following the same qualitative description of ionization-induced refraction in Chapter 3, we can readily derive that a necessary condition for a medium to guide intense laser pulses, instead of refracting them outward, is \( \partial n_r / \partial r < 0 \). Therefore, any mechanism that can contribute to constructing such radial distribution of refractive index is potentially beneficial for the intense pulse to propagate over a long distance.

The refractive index, which in essence modifies the phase and amplitude of the wave front, can be induced by several physical processes, including the electronic polarization, molecular orientation, electrostriction, saturated atomic absorption and thermal effects, although their response time scales may differ by orders of magnitudes. For example, the electronic polarization is usually regarded as instantaneous (~ \( 10^{-15} \) s), while it may take
as long as milliseconds for thermal effects to play a role, as summarized in Table 4-1 [4.12]. Therefore, when dealing with the propagation of subpicosecond pulses, we only take into account the electronic polarization and examine whether the induced refractive index has the favorable profile $\partial n_r/\partial r < 0$.

Table 4-1 Typical response time of the processes contributing to refractive index

<table>
<thead>
<tr>
<th>Physical processes</th>
<th>Electronic polarization</th>
<th>Molecular orientation</th>
<th>Electrostriction</th>
<th>Saturated atomic absorption</th>
<th>Thermal effects</th>
</tr>
</thead>
<tbody>
<tr>
<td>Response Time (s)</td>
<td>$10^{-15}$</td>
<td>$10^{-12}$</td>
<td>$10^{-9}$</td>
<td>$10^{-8}$</td>
<td>$10^{-3}$</td>
</tr>
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</table>

4.2 Mechanisms of Self-Channeling

4.2.1 Nonlinear self-focusing (Kerr effect)

Nonlinear self-focusing, or Kerr effect, is due to the dependence of the refractive index on laser intensities. This dependence originates from the third order susceptibility in the induced polarization field that can be written as:

$$P(t) = \varepsilon_0 \left[ \chi^{(1)} E(t) + \chi^{(2)} E^2(t) + \chi^{(3)} E^3(t) + \cdots \right]$$

(4-2)

For illustrating purposes we write the polarization and electric field as scalar quantities. $\chi^{(1)}$ to $\chi^{(3)}$ are the first- to third-order susceptibility and $\chi^{(2)}$ vanishes for materials with inversion symmetry, such as liquids, gases and certain crystals. Therefore, for the pulse propagation in gases, the refractive index that can be written as:

$$n(r) = n_0 + n_2 I(r)$$

(4-3)

where $n_0 = \sqrt{1 + \chi^{(1)}}$ is the familiar linear refractive index and:
From equation (4-3), we can see that for a Gaussian pulse \((\partial I(r)/\partial r < 0)\), the refractive index it feels has a negative gradient in radial direction \(\partial n(r)/\partial r < 0\), which is favorable in guiding the pulse. But this doesn’t automatically lead to the shrinking of a Gaussian pulse in the medium, since we also need to take into account the natural diffraction, which tends to spread the pulse. Qualitatively speaking, diffraction effect is inverse proportional to the square of the beam radius \(r \propto 1/r^2\), while Kerr effect is proportional to \(n_2 I\), i.e., \(n_2 P/r^2\), where \(P\) is the power of the pulse. Therefore, the same \(1/r^2\) dependence determines that if the Kerr effect overcomes diffraction at the beginning of the propagation, then the net effect of their competition is continuous focusing of the pulse until being terminated by some other nonlinear processes, e.g., stimulated Raman scattering, stimulated Brillouin scattering, two-photon absorption, or simply optical breakdown. Equating the diffraction with the Kerr effect thus gives us a critical power \(P_{cr}\) above which self-focusing occurs [4.13]:

\[
P_{cr} = \alpha \left( \lambda^2 / 4\pi n_0 n_2 \right)
\]

Here \(\alpha \sim 1.86\) is a constant that is independent of the material parameters. \(P_{cr}\) is sensitive to the wavelength of the pulse, as well as both the linear and nonlinear components of the refractive index. As an example, we plot in Figure 4-1 the critical power as a function of wavelength for the major components of air—Nitrogen \((n_2=2.2\times10^{-19} \text{ cm}^2/\text{W})\), Oxygen \((n_2=3.2\times10^{-19} \text{ cm}^2/\text{W})\), Argon \((n_2=2.0\times10^{-19} \text{ cm}^2/\text{W})\)—and the air itself \((n_2=2.4\times10^{-19} \text{ cm}^2/\text{W})\). The critical power is on the order of GW \((10^9 \text{ W})\), which is easily accessible within the delivering capability of our laser systems.
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The above analysis of the refractive index takes into account only the bound electrons. However, in almost all the applications that require extended propagation of intense pulses, ionization and thus free plasma electrons are prevailing. Non-relativistic refractive index of underdense plasmas can be written as:

\[ n(r) = n_0 + n_2 I(r) - \omega_p^2(r)/2\omega^2 \]  

(4-6)

As discussed in Chapter 3, the ionization-induced refraction following the plasma generation tends to refract the pulse outward. But for properly chosen parameters (laser power, plasma density, etc.), the defocusing introduced by diffraction and plasma generation can be canceled out by the Kerr effect, resulting in a self-guided optical beam (filamentation) with constant size and intensity. Filamentations can extend over many Rayleigh lengths, limited by energy depletion (e.g., due to ionization and collisional loss).

Figure 4-1. Critical powers of nonlinear self-focusing in major components of air
and some modulation instabilities. The clamping intensity and density of filamentations is far below the requirement from soft x-ray lasers, but this self-guided mechanism still leads to a lot of scientific discovery and practical applications, which are summarized in recent review papers [4.14]-[4.15].

4.2.2 Relativistic self-focusing

When the laser pulse approaches the relativistic regime, in which the quiver motion of electrons becomes relativistic, the dependence of the electron mass on the quiver velocity can result in a refractive index that is favorable for pulse guiding, i.e. $\partial n(r) / \partial r < 0$.

Before proceeding into details, a brief revisit to the laser strength parameter $a_0$ would be helpful. Defined as the peak amplitude of the normalized vector potential of the laser field, $a_0$ can be written in practical units as:

$$a_0 = eA/me^2 = 0.86 \times 10^{-9} \lambda[\mu m] \sqrt{I_0[W/cm^2]}$$ (4-7)

Or in terms of the laser electric field $E_L$:

$$E_L[TV/m] = 3.21a_0/\lambda[\mu m]$$ (4-8)

This reveals that the quiver velocity $v_q$, defined as $v_q = eE_L/m\omega$, is proportional to $a_0$.

Moreover, the relativistic factor $\gamma$ can also be written in terms of $a_0$ as:

$$\gamma(r) = \left(1 + a_0(r)^2 / 2\right)^{1/2}$$ (4-9)

Here $a_0=a_0(r)$ is to illustrate explicitly its spatial dependence. Equations (4-8) and (4-9) clearly show that $a_0$ can be used as a convenient parameter to characterize the intensity regime of a laser pulse, i.e. $a_0>1$ indicates a strongly relativistic pulse.
Taking into account the relativistic electron mass \( m_e = \gamma m_{e0} \) \((m_{e0} \text{ is the rest mass})\), the modified refractive index of underdense plasma is then:

\[
n(r) = 1 - \frac{\omega_{p0}^2}{2\omega^2\gamma(r)} \quad (4-10)
\]

Here \( \omega_{p0} \) is the non-relativistic plasma frequency. For a Gaussian pulse with \( \partial I / \partial r < 0 \), from (4-9) and (4-10), we can readily derive that \( \partial n(r) / \partial r < 0 \), which makes possible the self-guiding of the pulse.

**Figure 4-2.** Critical power of relativistic self-focusing in the plasma density range \( 10^{18} - 10^{21} \text{ cm}^{-3} \) at certain popular wavelengths (0.4 µm, 0.6 µm, 0.8 µm, 1.0 µm)

Following the same argument as in Kerr effect, we can conclude that there also exists a critical power \( P_{cr} \) for relativistic self-focusing [4.16]:

\[
P_{cr} = \frac{m_e c^5}{e^2 \omega_{p0}^2} = 17.4 \left( \frac{\omega}{\omega_{p0}} \right)^2 [\text{GW}] \quad (4-11)
\]
$P_{cr}$ is only dependent on the wavelength of the incident pulse and the density of plasmas in which the pulse is propagating. In Figure 4-2 we plot the critical power for pulses between 0.4 $\mu$m and 1 $\mu$m and plasma densities of $10^{18}$-10$^{21}$ cm$^{-3}$. In this parameter window, the critical power ranges from 20 GW to tens of terawatts, which are still accessible for our laser system. In relativistic channeling, electrons in the center of the pulse oscillate faster and thus become “heavier”, producing a larger refractive index than that on the fringe where laser intensity drops. The focusing effect from this refractive index profile can, in principle, balance the diffraction, as well as the defocusing from the plasma creation, thus initiating a self-channeling over a quite long distance.

However, two problems arise from this simplified physical picture. First, the modification of the refractive index requires a collective response of the plasmas. The time scale of this process is characterized by the plasma oscillation period ($\sim \omega_{p0}^{-1}$). Therefore, relativistic self-focusing becomes ineffective when the pulse duration is shorter than this time scale. Second, even for pulses that are long enough to establish the proper refractive index, they are still subject to instabilities such as self-modulation that are detrimental to the propagation [4.17]. Limited experimental reports on relativistic self-focusing can be found in [4.18]-[4.22].

4.2.3 Ponderomotive self-focusing

In addition to the quiver motion of electrons, changes of the spatial distribution of plasma density, induced by the ponderomotive force, can also create proper refractive index for pulse guiding. The ponderomotive force, defined as $F_p = -e^2/(4\omega^2)\nabla E^2$,
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describes the nonlinear force a charged particle feels in an oscillating electromagnetic field with inhomogeneous amplitude. The direction of the force, which is independent of the sign of the charge, is pointing toward the weak field area. Therefore, for a Gaussian pulse, its ponderomotive force expels both electrons and ions outward, although in different timescales ($\omega_{pe}^{-1}$ and $\omega_{pi}^{-1}$ for electrons and ions, respectively). This results in a suppression of the plasma density on the axis ($\frac{\partial n_e(r)}{\partial r} > 0$), translating into a favorable refractive index distribution ($\frac{\partial n(r)}{\partial r} < 0$), considering that $n = (1 - n_e / n_e)^{1/2}$.

Similar to relativistic self-focusing, the finite time required to build up the change of refractive index determines that the effectiveness of the ponderomotive self-focusing is also sensitive to pulse duration $\tau$, which at least has to be much longer than $\omega_{pe}^{-1}$. For $\tau \ll \omega_{pi}^{-1}$, ions will not have enough time to respond collectively. Under this stationary ions assumption, detailed simulation indicates that for pulses with less power than $P_{cr}$—threshold power for relativistic self-focusing—the induced channel depth, defined as the density difference between the axis and the edge of the plasma, is not deep enough for guiding [4.10]. As the power approaches $P_{cr}$, then the dominant guiding mechanism would be relativistic self-focusing, and the ponderomotive force can aid this guiding by slightly reducing the critical power to $0.93P_{cr}$ [4.16].

For pulses of moderate duration ($\omega_{pe}^{-1} \ll \tau < \omega_{pi}^{-1}$), the ion motion can be significant. The outward expelling of electrons sets up a large space-charge field which would drag the ions away from the axis. Ions continue drifting even after the extinction of pulses at the ion acoustic speed $C_s = (ZT_e / m_i)^{1/2}$ where $T_e$ and $m_i$ are electron temperature and ion mass respectively, and create a plasma channel. If the power is also sufficiently high
(\(P > P_{cr}\)), the combination of the relativistic and ponderomotive self-focusing can leave behind the pulse a high-density channel which is capable of guiding a second pulse. Krushelnick et al. observed the guiding of pulses with an intensity of \(5 \times 10^{16} \text{ W/cm}^2\) through a 2.5 mm long channel generated by a relativistic pulse of \(6 \times 10^{18} \text{ W/cm}^2\) \([4.23]\).

Some other mechanisms, such as pulse tailoring and plasma wave guiding, can also help the propagation of intense pulses, but in more subtle ways and they require more stringent conditions. They are described and reviewed in \([4.10]\).

### 4.3 Guiding of Intense Pulses in Plasma Waveguides

Plasma waveguides in essence are very similar to optical fibers that consist of a core surrounded by a cladding layer of lower refractive index, therefore, some general conclusions in optical fibers are also very useful here. Consider a gradient refractive index waveguide (in contrast to the step index waveguide whose refractive index jumps over the core-cladding interface) with the following structure:

\[
n(r) = n_0 - \frac{1}{2} n'' r^2
\]

(4-12)

where \(n_0\) is the refractive index of the fiber core and \(n''\) is a constant that characterizes the change of refractive index from the core to the cladding. For any guided mode, its amplitude modulus is independent on \(z\), \(i.e.\) the pulse propagates in the fiber with a constant intensity. Its general form can be written as:

\[
E(x, y, z, t) = E_0(x, y) \exp[i(\beta z - \omega t)]
\]

(4-13)
Here \( \beta \) is the longitudinal (in \( z \) direction) wave number of the guided mode. Substituting this trial solution and the refractive index distribution (4-12) into the wave equation (4-1) gives us:

\[
\left( \nabla^2_{\perp} - k^2 \frac{n^n}{n_0} r^2 \right) E_0 = \left( \beta^2 - k^2 \right) E_0 \quad (4-14)
\]

The above equation has the same form as the Schrödinger equation that describes a 2-D harmonic oscillator, with its eigenvalues replaced by the term \( \left( \beta^2 - k^2 \right) \). Its solution in cylindrical coordinates is:

\[
E(r, z, \phi, t) \propto e^{-r^2/w^2} r^m L_p^m \left( \frac{2r^2}{w^2} \right) e^{i\phi} \exp[i(\beta z - \omega t)] \quad (4-15)
\]

Here \( L_p^m \) is the Laguerre polynomial with \( p \) and \( m \) as its radial and azimuthal mode number. \( w \) is determined by the refractive index distribution and the vacuum wave number \( k \) of the pulse as:

\[
w = \left( \frac{2}{k} \right)^{1/2} \left( \frac{n_0}{n^n} \right)^{1/4} \quad (4-16)
\]

The dispersion relation of the modes satisfies:

\[
\beta^2 = k^2 - 2k \left( \frac{n^n}{n_0} \right)^{1/2} (2p + m + 1) \quad (4-17)
\]

For the lowest order mode \((p=0, m=0 \text{ and } L_0^0 = 1)\), the solution reduces to a simple Gaussian distribution with \( w \) as its width. This leads to an important matching condition: a Gaussian pulse with spot size \( w_M \) would excite only the lowest order mode and thus
propagate in the fiber with a constant transverse intensity profile, if its size matches the mode size, i.e. $w_M = w$.

Coming back to the plasma waveguides, the establishment of the refractive index profile as in (4-12) is usually realized by a parabolic plasma density distribution:

$$n_e(r) = n_e(0) + \Delta n_e \left( \frac{r}{r_{ch}} \right)^2$$

Here $r_{ch}$ is the radius of the plasma waveguide and $\Delta n_e = n_e(r_{ch}) - n_e(0)$ is the channel depth.

The refractive index, assuming that the plasma is underdense, is thus:

$$n(r) = 1 - \frac{1}{2} \frac{n_e(0)e^2}{m_e c \omega^2} - \frac{1}{2} \frac{e^2}{m_e c \omega^2} \Delta n_e \left( \frac{r}{r_{ch}} \right)^2$$

Comparing (4-19) with (4-12) gives us:

$$n'' = \frac{e^2}{m_e c \omega^2} \Delta n_e$$

Moreover, $n_0$ is taken as unity here to the zero-order approximation in underdense plasmas. So the matched spot size for a plasma waveguide with radius $r_{ch}$ and depth $\Delta n_e$ can be readily derived:

$$w_M = \left( \frac{r_{ch}^2}{\pi r_{ch}^2 \Delta n_e} \right)^{1/4}$$

Here $r_e = (1/4\pi\varepsilon_0)(e^2/m_ec^2)$ is the classical radius of the electron. The matched spot size for a given plasma waveguide is independent of the pulse wavelength. Therefore, plasma waveguides not only guide the ultra-short (~ 100 fs) and ultra-intense (~ $10^{18}$-$10^{19}$ W/cm$^2$) pump pulses, but also help the propagation of the soft x-ray radiation, reducing the
refraction losses that used to set a severe limit on the usable density and length of plasmas in soft x-ray lasers (see e.g. [1.28]).

In practice, plasma waveguides can be formed with several techniques, out of which the hydrodynamic expansion following the ionization of a line-focused laser pulse is of particular interest. Pioneered by Milchberg and his group [4.25], this method first creates a spark in gases in the line focus of an axicon lens. Ion-ion and ion-atom collisions during the plasma expansion results in a shockwave front propagating in radial direction toward the surrounding neutral or weakly ionized gases. A plasma channel is then formed behind the shock wave, with density minimum on the axis and maximum in the shock front. The back-filled gases were later replaced by gas jets which were more suitable in practical applications [4.26]. Several two-pulse techniques were also investigated. The transmission of the guided pulse was enhanced by increasing the height and thickness of the waveguide’s density barrier using two-pulse-excitation method [4.27], in which both pulses were 100 ps. The igniter-heater technique using cylindrical lenses [4.28] or axicons [4.29] was later developed. In this scheme, a short pulse (igniter, <100 fs) produces a small concentration of seed electrons in the gas through multi-photon ionization or field ionization. The relatively longer pulse (heater, 80-160 ps) arrives after a certain delay and efficiently heats up the pre-formed plasma through the collisional absorption and drives the formation of plasma waveguides. The absorption of the laser energy that drives the plasma expansion is usually low, typically on the order of 10%. This issue was addressed by creating the plasma channel in gas clusters [4.30], which are atomic or molecular assembly bonded by van der Waals forces and can be formed by following the rapid cooling during the expansion of gases through a nozzle. The particle
density within each cluster is extremely high—comparable to solids—so the collisional heating is rapid and efficient.

The pre-formed plasma waveguide method distinguishes itself from the variety of self-channeling mechanisms by its superior control over the waveguide properties. Since the waveguide is created by another, independent laser pulse, its diameter, length, channel depth and on-axis density can be adjusted to cover a wide parameter range and satisfy requirements from practical applications. This advantage constructs the basis of our preference on plasma waveguides in extending the propagation of the ultra-intense pump pulse in soft x-ray lasers.

4.4 Formation of Large Diameter Plasma Waveguides

4.4.1 Experimental setup

We investigate here the plasma formation using the modified igniter-heater technique in which a 200 ps pulse was employed as the igniter, followed by a 10 ns heater propagating at the same direction, as shown in Figure 4-3. Two laser systems, synchronized electronically, have been used in this experiment. A Quanta-ray Nd:YAG laser with output pulses of 600 mJ, 10 ns, 1064 nm was employed as the heater pulse, while the igniter pulse was provided by the Thales Alpha system (200 ps, before compression). The 200 ps igniter pulse created plasma channels in the line focus of an axicon after passing through the mirror for 1 \( \mu \)m wavelength. The axicon, manufactured by Asphericon, Germany, had a 20° base angle and ~46 mm long line focus. The peak on-axis intensity point, 30 mm away from the tip of the axicon, was determined
experimentally by gradually decreasing the input energy until a minimal filament was achieved. Intensities at this point were $1.2-2.4\times10^{13}$ W/cm$^2$, corresponding to 100 -200 mJ input energies. Due to the small intensity gradient around peak point, we assumed a uniform intensity along the 6 mm long orifice of the gas jet. The number of concentric rings in the Bessel beam could be estimated geometrically. The radius of the optical cone at the plasma was 2.74 mm. Rings are of equal spacing $\Delta R=2.34\ \mu m$, giving a total number of $\sim1100$ rings. The heater pulse was focused by an $f=60$ cm lens (F/30) into the entrance of the plasma channel created by the igniter after passing through the central hole ($\sim5$ mm diameter) of the axicon. Rayleigh length of 2.7 mm then can be expected under this condition and it was between the thickness ($\sim1$mm) and length ($\sim6$mm) of the gas target.

Figure 4-3, Experimental setup for the modified igniter-heater technique in creating large diameter plasma waveguides
The plasma was created in a supersonic gas jet using a pulsed gas valve as described in Chapter 3. We used a rectangular orifice of $6\times0.5$ mm$^2$, which allowed us to change the effective depth of the gas target by changing the orientation angle of the orifice with respect to the laser propagation axis. The neutral gas density was controlled by changing the back pressure or its opening time. Two orientations of the orifice have been used. In one orientation, the laser pulse propagated across the orifice in order to monitor the gas and plasma density simultaneously. In the other orientation, when the pulse was along the orifice, we could create long and uniform plasma channels. All the data presented here are in transverse orientation unless otherwise specified.

Plasma diagnostics were performed by a shear-type interferometry in which the interference pattern was produced by overlapping two parts of a single collimated beam provided by the Positive Light laser system (200 fs, a few mJ). The signal part passed through the plasma region while the reference part bypassed the gas jet and carried the reference phase. A biprism and collimating lenses recombined them on a CCD, which was protected from plasma radiation by a 400 nm narrow band-pass filter. Absolute measurements of phase shift on a shot-to-shot basis were possible due to the robustness of the system against mechanical and acoustic noises. Temporal and spatial resolution was 200 fs and 5 $\mu$m, determined by the duration of the probe pulse and the numerical aperture of the imaging system, respectively. An optical delay line with 50 $\mu$m step size (100 $\mu$m of a round trip for the probe) was installed to adjust the delay between the probe and the two ionization pulses, enabling us to observe the evolution of plasmas with a time step of $\sim 0.33$ ps.
4.4.2 Characterization of the axicon lens

One of the key elements in this setup is the axicon lens, which has a conical front surface that focuses the incident beam into a longitudinal line. The axicon lens employed in our experiment, as specified in Figure 4-4, is made of fused silica, which has a refractive index \( n \approx 1.45 \) at 800 nm. Straightforward analysis in geometric optics can reveal a few important facts about the axicon lens. From Snell’s law \( \sin(\beta + \alpha) = n \sin(\alpha) \), where \( \alpha = 20^\circ \) is the base angle of the axicon, the half angle \( \beta \) of the focal cone can be readily derived as \( 9.73^\circ \). The incident beam has 16 mm diameter and its central part of 5 mm diameter directly passes through the hole in the center of the axicon without contributing to the line focus.

![Figure 4-4, Specification of the axicon lens used in our experiment](image)

The length of the line focus (the shadow area in Figure 4-4) is about 32 mm, calculated from \( L_0 = (R_0-r_0) / \tan(\beta) \) where \( R_0 = 8 \) mm and \( r_0 = 2.5 \) mm are the radius of the beam and the hole, respectively. For any small section \( z \)-\( z + \text{d}z \) of length \( \text{d}z \) on the line focus, its energy comes only from a narrow ring between radius \( r_\beta \) and \( r_\beta + \text{d}r_\beta \) of an area \( 2\pi r_\beta \text{d}r_\beta \) on the incident beam, where \( r_\beta = z \tan(\beta) \). In another word, \( I(z)\text{d}z \propto I(r_\beta)2\pi r_\beta \text{d}r_\beta \), where \( I(r_\beta) \) is
input beam intensity at $r_\beta$. For a Gaussian input beam, the intensity drops as the radius increases, but at the same time the area of the contributory ring also becomes larger. Their competition results in a peak intensity point on the line focus, which is approximately 30 mm away from the tip of the axicon. We marked this peak intensity point in red in Figure 4-4, but exaggerated its length for illustrating purpose.

The scalar theory of diffraction, which is a good approximation for paraxial rays of any polarization, shows that the pulse intensity at any point $(y, z)$ in the focal region of an axicon lens can be written as [4.31]-[4.33]:

$$I(y, z) = I(r_\beta) \left( \frac{2\pi k r_\beta \sin \beta}{\cos^2 \beta} \right) J_0^2(ky \sin \beta)$$

(4-22)

where $J_0$ is the zero-order Bessel function of the first kind, $k$ is the wave number, and $y$ is the transverse distance from the axis (see Figure 4-4). Two interesting features arise from this solution. First, for any cross section in the focal region, the transverse intensity profile is a $J_0$ function, which has a central maximum and equally-spaced concentric rings extending to infinity. In reality, due to the limited aperture of the axicon, the Bessel distribution also has a limited size (~2.74 mm here) as can be seen from Figure 4-4. For points on the axis, i.e., $y=0$ and $J_0(0)=1$, the intensity is proportional to $I(r_\beta)r_\beta$ which is in agreement with our geometrical analysis.

Second, the radius of the beam at focus, defined as the distance between the center and the first zero in Bessel function, is independent on $z$, i.e. it maintains a constant beam size over the whole focal region:

$$y_0 = 2.408 / (k \sin \beta)$$

(4-23)
The spot size is only a function of the wave number $k$ and the angle of focal cone $\beta$. In our case, $\lambda=800 \text{ nm}$ and $\beta=9.73^\circ$, so the generated Bessel beam has a constant spot size $D=3.6 \mu\text{m}$ over the focal line with a length $L_0=32 \text{ mm}$. In comparison, a Gaussian beam of the same spot size and wavelength has a Rayleigh length of merely 50 $\mu\text{m}$ long, almost three orders of magnitude shorter than $L_0$. As a result, Bessel beams are usually called diffractionless beams as though it overcomes the natural diffraction.

![Figure 4-5](image)

**Figure 4-5.** (a) original CCD image of the cross section of the Bessel beam and (b) its radial intensity distribution

To test the quality of our axicon, we illuminated it with a collimated He-Ne beam ($\lambda=632.8 \text{ nm}$) and observed the beam profile at the focus with a CCD camera. Figure 4-5 (a) shows us the concentric rings of the intensity profile. The measured size of the central spot is 2.94 $\mu\text{m}$, in excellent agreement with the theoretical value of 2.82 $\mu\text{m}$. We then extracted the radial profile and fit it with $I(y)=pJ_0(ky\sin\beta)$, where $p$ is a fitting parameter. Very good agreement was achieved, as seen in Figure 4-5 (b). Equation (4-22) is used to compute the peak intensity of the Bessel beam. For our igniter, $\lambda=800 \text{ nm}$, pulse duration $\tau=200 \text{ ps}$, beam diameter $D_0=16 \text{ mm}$, intensities of $1.2-2.4\times10^{13} \text{ W/cm}^2$
can be achieved at around 30 mm away from the tip of the axicon, with moderate input energies of 100-200 mJ.

It is worth to mention here the importance of good alignment in achieving zero-order Bessel beam from axicon lens. The incident beam needs to be in the center of the axicon aperture and perpendicular to its flat back surface. Slight misalignment would result in a distorted focus [4.34] and thus destroys the cylindrical symmetry of the plasmas. We employed a He-Ne beam passing through the center of an iris diaphragm to assist the alignment. The aperture of the diaphragm was set to be slightly larger than the hole in the axicon, so part of the beam was reflected back by the flat surface of the axicon. By adjusting the last reflection mirror before axicon and tilting the axicon itself, we could: 1) make the reflection a full and symmetrical ring, which indicates that the incident beam is in the center of the axicon; and 2) propagate the reflection following exactly the same path as the incident beam but in reverse direction to confirm the perpendicularity. Then we overlapped our igniter pulse with the He-Ne beam to establish the good alignment. We cannot directly observe the reflection of the igniter beam due to the anti-reflection coating at 800 nm on the flat surface of the axicon.

### 4.4.3 Plasma creation using the Bessel beam only

Figure 4-6 shows a photo of a typical plasma column created by the Bessel beam in air. The plasma is comparable in length with $L_0$, the extent of the focal region. The starting point of the plasma $z_0=15$ mm is estimated by $z_0=r_0/\tan(\beta)$, where $r_0$ is the radius of the center hole and $\beta$ is the half angle of the focal cone. A sample interferogram of the igniter-created plasma is shown in Figure 4-7. The plasma is created in a chamber filled
with nitrogen at 500 Torr and it is observed 2 ns after the igniter pulse. Limited by the field of view of the imaging system, we only show a portion of the plasma. Throughout the entire length, the plasma maintains a constant size, confirming the diffractionless nature of the Bessel beam. The thickness of the shock wave front is on the order of the mean-free-path of ion-ion collisions $\lambda_{i,i}$. For nitrogen at density $N_i = 10^{19}$ cm$^{-3}$, $\lambda_{i,i}$ is $\sim 0.25 \mu$m [4.11], which is beyond the resolution of the imaging system. So, a sharp discontinuity on the fringe appears at the position of the shock wave front, leaving a clearly defined boundary of plasma channels. Throughout this chapter, we use this position to mark the diameter of the plasmas.

**Figure 4-6, A typical plasma column created by Bessel beam in air**

The expansion of the igniter-created plasmas represents a typical example of a self-similar flow, in which the variables change with the time in such a manner that their distribution with respect to the coordinate variables always remains similar in time. The self-similar solution of a cylindrical blast wave is [4.35]:

$$R(t) = \frac{\xi}{s_0} (E_{th} / \rho_0)^{1/4} t^{1/2}$$

(4-24)
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Here $R(t)$ is the distance between the shock wave front and the axis, $E_{th}$ is the thermal energy per unit length that drives the expansion, $\rho_0$ is the initial mass density, and $\xi_0$ is the dimensionless parameter which is a function of the specific heat ratio and is on the order of unity.

Figure 4-7, Interferogram of plasmas created by igniter (104 mJ, 800nm, 200 ps) in chamber filled with N$_2$ at 500 Torr, delay between probe pulse and igniter is 2 ns

Figure 4-8 shows the expansion of plasmas created in N$_2$ jet using different energies and the fitting of experimental data points with $D(t)[\mu m] = p_1(t[ns] + p_2)^{p_3}$ using least-square method. $p_1$ to $p_3$ are fitting parameters: $p_1$ is the $\xi_0(\sqrt{E_{th}/\rho_0})^{1/4}$ factor in equation (4-24), $p_2$ suggests the exact moment of plasma creation and $p_3$ determines the dimension of the expansion ($p_3=0.5$ for cylindrical blast wave and 0.4 for spherical one) respectively. For the three pulse energies—104 mJ, 148 mJ and 196 mJ—we use here, $p_1$ is 61.3, 72.5 and 83.6 respectively, and roughly follows the $E_{th}^{1/4}$ dependence assuming that a fixed
portion of the pulse energy is deposited into the plasma to drive the expansion. \( p_2 \) is 24±2 ps for all three cases, indicating that the plasmas are initiated 24 ps before the peak of the igniter pulse. Also for all the three cases, the corresponding \( p_3 \) is 0.54, 0.51 and 0.50, all within 10% of 0.5, and thus verifies the cylindrical symmetry of the plasma, an important condition under which we can safely use the inverse Abel transform to extract the radial plasma density from interferograms.

![Figure 4-8](image)

**Figure 4-8, Expansion of plasma channels created by the Bessel beam in \( N_2 \) gas jet**

The expansion speed, calculated by taking the derivative of the fitted curve \( R(t) \) \((R=D/2)\) is plotted in Figure 4-9. The initial expansion speed immediately after the extinction of the laser pulse is the range of 4.5-7×10⁶ cm/s. As the plasma expands, the speed quickly drops to less than 10⁶ cm/s after 2-2.5 ns. The near-saturation behavior of the expansion speed at longer delays (>2ns) makes it possible to extrapolate the plasma size at delays beyond the time span used in this experiment and thus design proper timing.
for the injection of subsequent pulses. During the delays considered here, the plasma expansion is always supersonic. For ideal nitrogen at a room temperature $T=300 \, K$, the estimated speed of sound is close to $3.5 \times 10^5 \, \text{cm/s}$ ($c_s = \sqrt{\gamma kT/m}$), where $\gamma=1.4$ the heat capacity ratio for diatomic gases. This agrees with the physical depiction of the plasma expansion as the propagation of shock wave front.

![Figure 4-9](image)

**Figure 4-9,** Expansion speed of the nitrogen plasmas derived from the fitted expansion curve $R(t)$

To investigate the guiding property of the plasmas created by the Bessel beam, we have derived the radial plasma density using the inverse Abel transform and compiled the density profiles at different probe delays in Figure 4-10. Density suppression on the axis appears almost immediately after the passage of the pulse (starting from 0.2 ns). As the plasma radius, defined as the distance between the density peak and the axis, expands from initially less than $10 \, \mu m$ to more than $65 \, \mu m$ after 4 ns, the peak density drops
correspondingly, from more than $4 \times 10^{19}$ cm$^{-3}$ to less than $0.5 \times 10^{19}$ cm$^{-3}$. Nevertheless, the on-axis density suppression persists, maintaining the guiding property of the plasma column.

![Figure 4-10](image1) **Figure 4-10**, *Radial density of nitrogen plasmas created by a Bessel beam of 196 mJ*

![Figure 4-11](image2) **Figure 4-11**, *Expansion of plasmas created by Bessel beam in (a) H$_2$ and (b) C$_2$H$_6$ jet*

We have also performed similar experiments in the hydrogen and ethane jet, with the results presented in Figure 4-11, Figure 4-12 and Figure 4-13. Hydrogen and ethane
plasmas have similar expansion with the incident pulse of 196 mJ. However, the size of the hydrogen plasma is more sensitive to the incident energy: at 4 ns with 148 mJ energy, the hydrogen plasma expanded to about 130 \( \mu m \) while the ethane plasma has already approached 160 \( \mu m \). This distinction is more clearly seen by the expansion speed shown in Figure 4-12. Expansion speed of ethane plasmas using different pulse energies almost all converged to \( 10^6 \) cm/s at 4 ns, but at the same delay hydrogen plasmas are still expanding at quite different speeds (50% difference between 148 mJ and 196 mJ). The radial density profiles are shown in Figure 4-13. Ethane plasmas already establish the guiding structure at the first observation point. Notice that although this point is marked by “0 ns” in the legend, the plasma is very likely to be created somehow 50 ps earlier implied by the \( \rho_2 \) value in the fitting of the expansion curves. In comparison, it takes the hydrogen plasma much longer (>0.5 ns) to form the density suppression on the axis, and within a shorter period of time the density drops below \( 10^{19} \) cm\(^{-3} \). These features directly influence the injection scheme for a second pulse in order to satisfy the mode matching conditions.

![Figure 4-12](image). Expansion speed of plasmas created by Bessel beam in (a) \( H_2 \) and (b) \( C_2H_6 \) jet at different energies.
Plasmas created by a single Bessel beam have been successfully employed as waveguides for powerful beams since the beginning of the experimental realization of this concept. For some application such as Raman amplification, the pulse to be guided is often quite wide ($D>200 \, \mu m$) in order to keep the intensity below wave breaking threshold without sacrificing the pulse energy. Moreover, the resonant condition for the Raman backscattering process requires the plasma waveguide to maintain a density close to $1.3 \times 10^{19} \, \text{cm}^{-3}$. As seen from the above results, the size and density requirement cannot be simultaneously satisfied in plasmas created by a single Bessel beam: either the plasma is too narrow when the density is high enough, or the density drops too low when the plasma is sufficiently wide.

In order to resolve this contradiction, we proposed to inject a second pulse of nanosecond duration as a heater. In this scheme, the 200 ps Bessel beam created a preliminary waveguide to guide through the ns pulse, which further ionized the plasma and deposited more energy through collisional heating such that it drove the expansion more effectively, producing a final waveguide that was both wide and dense enough for practical applications.
4.4.4 Injection of the nanosecond pulse

The 10 ns heater pulse, delivered by an Nd: YAG laser of Spectra Physics Quanta-Ray series, had 600 mJ at 1.064 µm and it was focused by a spherical lens (f=60 cm) into a 40 µm wide spot at the entrance of the preliminary plasma waveguide generated by the Bessel beam. Although a single spherical beam was used in creating plasma waveguides before (see, e.g. [4.25]), a series of nonlinear processes, particularly the ionization-induced refraction, prevented it from propagating a distance longer than its Rayleigh length. And the created plasmas, as shown in Figure 4-14 (a), suffered from serious modulation in both size and density. Also, from shot to shot, the plasmas were irreproducible, rendering its practical application very unreliable.

![Figure 4-14](image)

**Figure 4-14.** Plasmas created in the ethane jet using (a) a single spherical beam, 10 ns, 600 mJ, 1.064 µm; (b) a single Bessel beam, 200 ps, 196 mJ, 800 nm; and (c) the combination of spherical and Bessel beams, with 8 ns delay between the two pulses, probe beam is 11 ns after the peak of the Bessel beam. Both ionization beams are along the gas jet.
In Figure 4-14 (b) we present the plasmas created by the single Bessel beam 11 ns after the passage of the pulse. Although the plasma maintains uniform, its density was already way below $10^{19}$ cm$^{-3}$ as we discussed in last section. It seems that each individual pulse had certain limitation in fulfilling our purpose. However, when we combined the two pulses together, with careful alignment, these individual limitations were cleanly eliminated, resulting in plasma waveguides of satisfactory quality. The spherical beam propagated, and ionized, all the way through the preliminary plasma waveguide, while driving its expansion effectively and uniformly in the radial direction.

![Figure 4-15](image_url)  

**Figure 4-15.** Expansion of plasmas created in nitrogen jet using a single Bessel beam ('*') and the Bessel-Spherical combination ('x'); the Bessel and Spherical beam has 148 mJ and 600 mJ, respectively, with 0 ns delay between them

Again we start our quantitative investigation of plasma waveguides by examining the expansion curves due to their close relationship with the efficiency of energy deposition. Comparison of the plasma expansion is presented in Figure 4-15, which reveals a
dramatic enhancement of plasma diameter after the injection of the heater pulse. The fitting parameter $p_1$ turns out to be 60 for the single pulse case and 212 for the combination of two pulses. Considering the biquadratic dependence of $E_{th}$ on $p_1$, our injection of four times more energy (600 mJ of the heater pulse over 148 mJ of the igniter pulse) yields over two orders of magnitude enhancement in energy deposition ($[212/60]^4$)!

More importantly, the energy from the heater pulse is distributed uniformly along the plasma to drive the expansion as can be seen from the original interferogram. Notice that the $p_2$ value is -6.4 ns in this case, which suggests that the heater pulse doesn’t start its effective interaction with the preformed plasma until 6.4 ns after the peak of the Bessel beam. By effective interaction we mean that the heater pulse starts depositing a significant amount of energy into the plasma and noticeably changes the way it expands. In another word, 6.4 ns marks a critical point after which the plasma expansion deviates its original track (dashed line) and starts following a new trajectory (dotted line) as seen in Figure 4-15.

**Figure 4-16.** Expansion of ethane plasmas using the Bessel-Spherical combination with different energies of Bessel beam
Figure 4-16 presents the expansion of two plasma columns using different igniter energies and it can be seen that the igniter energy plays little role in the plasma expansion. So, unlike the plasmas created by a single Bessel beam as shown in Figure 4-8, the energy to drive the expansion here in the igniter-heater scheme is mostly from the heater pulse. This is an advantage in experiments since it is usually much less expensive to scale up the pulse energy from nanosecond lasers comparing to picosecond lasers.

![Figure 4-17](image)

**Figure 4-17.** Average density of plasmas created in nitrogen jet in comparison with the neutral gas density; both laser pulses are across the jet, 148 mJ for igniter and 600 mJ for the heater; two igniter-heater delays—0 ns (‘*’) and 4 ns (‘x’)—are used.

After confirming that more energy is deposited into the plasma to drive the expansion, we then investigated whether additional ionization also occurred to improve the overall density of the widened plasmas. We first shot both pulses across the gas jet in order to monitor simultaneously the neutral gas density and the plasma density, and studied their relationship. Then we rotated the orifice of the jet by 90° to make it parallel to the laser pulses, aiming to elongate the plasma column.
In Figure 4-17 we present the average plasma density in comparison with the molecular density of neutral gas in nitrogen jet. A striking feature of these curves is that the plasma density distribution almost exactly follows the gas density distribution. In another word, the plasma has a constant ionization stage along the column, which indicates that the heater pulse maintains a constant intensity when propagating in the plasma. Therefore, if we deliver the laser pulses along the gas jet in which case they interact with a uniform gas target of 6 mm long, it is reasonable to expect a uniform plasma density along the channel. Figure 4-18 shows the results. Throughout a length of about 3 mm, the plasma columns maintain an average density around $2.4 \times 10^{19}$ cm$^{-3}$, within $\pm 15\%$ variation. Notice that this 3 mm column is only the section that falls into the field of view of our imaging system. The real plasma column is even longer and similar uniformity can reasonably be expected.

![Figure 4-18](image)

**Figure 4-18.** Average density of plasmas created in nitrogen jet, laser pulses are along the gas jet, 196 mJ for igniter.
These plasma columns also show certain tolerance over the delay between the igniter and the heater. In fact, the igniter-heater delay determines the size and density profile of the preliminary plasma waveguide created by the Bessel igniter at the moment when the heater arrives. Hence, within a few nanoseconds, the preliminary waveguides are all capable of guiding the heater pulse for additional ionization and expansion. This tolerance is remarkable. Figure 4-19 shows the average density of plasmas created by the igniter-heater technique with an igniter of 148 mJ. Although we observe no significant visual difference in the original interferograms, quantitative analysis reveals much more significant variation in the axial density. In practical applications, any method that can control the plasma waveguide profile would be beneficial in optimizing the parameter window for pulse guiding. Methods at our disposal include gas density, focal position of
the guided pulse, igniter energy and the igniter-heater delay, out of which the last two are most frequently used. Changing the gas specie, in applications that don’t require specific ions such as the Raman amplification, is also a helpful method and therefore we have explored the plasma columns in ethane jets as well.

![Figure 4-20](image)

**Figure 4-20.** *Average density of plasmas created in ethane jet, laser pulses are across the jet, various igniter energies have been used*

In contrast to nitrogen plasmas, ethane plasmas are quite robust to the variation of the igniter energy as can be clearly seen in Figure 4-20. Almost no visible change of plasma density is induced when we tune the igniter energy between 196 mJ and 104 mJ. And with all the three energies we use, the plasmas maintain an impressively steady charge state along the pulse propagating direction. On average, one electron is stripped from each ethane molecule. This is because the plasma density is mostly contributed by the ionization of the heater pulse (the density of the igniter-created plasma is very low when the heater arrives, see Figure 4-10) and its energy is fixed in our experiments.
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In Figure 4-21, we compile the distribution of average plasma density with two sets of igniter energies and igniter-heater delays when the pulses are traveling along the gas jet. For each specific set of parameter, the density variation is within 10% over a length of 2.5 mm that is picked up by the imaging system. The absolute density fluctuation is approximately \(\pm 3 \times 10^{18} \text{ cm}^{-3}\), comparable to the resolution of the interferometric diagnostic method that is \(1.2-2.5 \times 10^{18} \text{ cm}^{-3}\), corresponding to a quarter fringe shift over a plasma breadth of 200-400 \(\mu\text{m}\). Even the overall density fluctuation in this chosen parameter window is only \(\pm 0.6 \times 10^{19} \text{ cm}^{-3}\), or 18%, and should satisfy most of the uniformity requirement imposed by practical applications.

![Figure 4-21](image)

**Figure 4-21**, Average density of plasmas created in ethane, laser pulses are along the gas jet, two sets of igniter energies and delays are used.

The unfolding of the radial plasma density is presented in Figure 4-22. We also compare the different density profiles of nitrogen and ethanem plasmas using the same
laser parameters. Qualitatively, density rise from minimum to the peak in ethane plasmas is much steeper than that in ethane. The ethane plasma extends 50 $\mu m$ further between the dip and the peak of the plasma density, although both of them are terminated at the same radius $\sim 275 \mu m$.

![Figure 4-22](image)

**Figure 4-22**, Radial density distribution of plasmas created in nitrogen and ethane jet, 196 mJ for igniter pulse, 600 mJ for heater pulse, 8 ns between them, 11 ns probe delay, ionization laser pulses are along the gas jet

This distinction could be ascribed to the different traces of the fringe bending as depicted in Figure 4-23. In both cases, the net bending of fringes from their background straight lines that are outside the plasma area is toward the left. This is determined by the arrangement of imaging system and is independent of the plasmas. However, the paths they take are quite in contrary. The nitrogen plasma displays a smooth and monotonic fringe transition from the background to the bending, while two sharp turns occur on the fringes of the ethane plasma. Sharp turns in fringe trace result in likewise phase-shift profile and thus steep density gradient occurs correspondingly.
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Figure 4-23. Bending of fringes in interferograms of (a) N2 and (b) C2H6 plasmas, shape of the bending is highlighted in white dashed line

One possible explanation of the sharp turns in the ethane plasma fringes could be the existence of a series of molecule ions such as C\textsubscript{2}H\textsubscript{6}\textsuperscript{+}, C\textsubscript{2}H\textsubscript{5}\textsuperscript{+}, etc. accompanying the ionization of C\textsubscript{2}H\textsubscript{6} [4.36]. These ions have very complex contribution to the total refractive index and could shift the fringes in complicated ways, instead of a smooth curvature. More detailed analysis is probably needed to reveal a more conclusive origin of this phenomenon. Nevertheless, the dependence of plasma density on gas species gives one more parameter through which we can further customize our plasma waveguide design. This igniter-heater method is now employed as an essential component in reaching high efficiency amplification in Raman backscattering.

4.5 Formation of Small Diameter Plasma Waveguides

4.5.1 Experimental setup

Unlike the Raman amplification, which requires large diameter pump beams, the soft x-ray laser benefits from tightly focused pulses ($r\lesssim10 \, \mu m$) which can lead to extremely
high intensity. The created small diameter plasma channel can also help the escape of energetic electrons that would otherwise impose negative effects on the creation of population inversion. In order to generate these small diameter waveguides, we set up another igniter-heater system as shown in Figure 4-24.

![Figure 4-24](image)

**Figure 4-24.** *Experimental setup for the creation of narrow plasma waveguides using the combination of 100 fs igniter and 200 ps heater*

In this setup, the p-polarized, 100 fs igniter pulse delivered by the Alpha Thales system, and the s-polarized, 200 ps heater pulse that was also from the Thales but uncompressed, were combined using a thin film polarizer. The plasma channel was formed after the optical breakdown of gases in Bessel-like radial intensity distributions created through passing both beams through the axicon lens. The length of the plasma channel formed with the axicon reached about 3.2 cm with current input beam diameter of 16 mm. Time-resolved plasma density distributions were measured with an interferometer using a 200 fs, 400 nm laser beam as the probe. In order to resolve the radial profiles of the plasma
columns with extremely small diameters, a long working distance, infinity-corrected 10× objective lens (NA=0.28) was used in the imaging system to improve its resolution to \(\sim 1 \mu m\). The interference fringes were formed as following: the plasma image was first split into two images through a beam-splitter; then they were shifted with respect to each other after being reflected from two mirrors and formed the interference pattern on the imaging plane. The interferograms were recorded in a single-shot mode using a 1280×1024 CCD camera. Separation of the two images of the plasma on each interferogram was limited by the density of fringes that could be distinguished by the detector. In our case, the separation was no larger than 50 \(\mu m\) on the object.

4.5.2 Corrugation in the Bessel beam-created plasma channels

Using the imaging system with improved resolution, we have observed prominent axial modulations in the plasma columns created by a single Bessel beam as shown in Figure 4-25. The optical breakdown initially creates a series of plasma bubbles along the axis and they expand in both radial and axial directions until merging into a uniform plasma column. The time scale for the merging to complete is usually on the order of nanoseconds depending on the expansion speed and the spacing between bubbles.

In Figure 4-26, we plot the expansion of the plasma bubbles up to 2 ns when they merge into a single channel and fit the experimental points to \(D = p_1(t - p_2)^{p_3}\) using least-square method. The best \(p_3\) value returned from fitting is 0.385, which is quite close to 0.4 that characterizes the propagation of spherical blast waves [4.35]. This set of data is taken when we focus the Bessel beam into the chamber filled with 1 atm nitrogen gas.
We also observed very similar behavior in air and gas jets using N\textsubscript{2} and C\textsubscript{2}H\textsubscript{6}, all of which had the best $p_3$ value within 10% of 0.4.

**Figure 4-25.** Evolution of plasmas created by a single 200 ps Bessel beam in chamber filled with 1 atm N\textsubscript{2} gas, 300 mJ pulse energy, the probe delay from top to bottom is 0.1 ns, 0.2 ns, 0.3 ns, 0.5 ns, 1 ns and 2 ns.

For further investigation, we examined the influence of incident pulse energy on the plasma bubbles in N\textsubscript{2} gas jet and presented the results in Figure 4-27 and Figure 4-28. Reduction of the pulse energy results in a shorter plasma since the low density area at the edge of the gas jet requires high pulse intensity for breakdown. But the modulation wavelengths, defined as the distance between the bubbles, almost maintain a constant
value (22-24 $\mu$m) regardless of the incident energy. The plasma diameters, chosen at the center of the gas jet, as a function of incident pulse energy are plotted in Figure 4-28. The experimental points are again fitted to $D = p_1(E - p_2)^{p_3}$, where $E$ is the pulse energy in $mJ$. The returned $p_3$ value is 0.188, which is in agreement with the $E^{1/5}$ dependence of spherical blast waves. Combining Figure 4-26 and Figure 4-28, it seems that the initial plasmas, prior to the formation of a single uniform column, are following the self-similar solution of a spherical blast wave.

![Fitting curve](image)

**Figure 4-26**, Expansion of plasma bubbles created by a single 200 ps Bessel pulse in quiet N$_2$ gas, 300 mJ pulse energy

A noticeable feature of the plasmas in gas jet is its spindle-shaped envelope of the plasma bubbles as illustrated by the red dashed line in Figure 4-29 (a). The bubbles have largest sizes in the center of the jet and gradually shrink toward the edge. Figure 4-29 (b) presents the plasma-bubble diameters which have a quite good fitting to a Gaussian
distribution that is also the shape of the gas density profile across the jet. A few counteractive processes that are related to the local gas density might lead to this observation. The plasma expansion is proportional to $(E_t / \rho_0)^{1/5}$ for spherical blast waves. On the one hand, any increase of the initial gas density would slow down the plasma expansion through the $(1/\rho_0)^{1/5}$ factor. On the other hand, however, higher gas density results in more frequent electron-ion collisions and thus more efficient collisional heating that would drive the expansion faster through the $E_t^{1/5}$ factor. These two processes tend to cancel out each other, so the third process becomes decisive. Regions of different initial gas densities have different threshold intensities for breakdown. The center of the jet is probably ionized at the beginning of the pulse while the ionization on the edge occurs upon the arrival of the peak intensity of the pulse. Therefore, upon the arrival of the probe beam, plasmas in the center expand for a longer time than those on the edge, resulting in the observed inhomogeneous plasma size.

![Interferograms of plasmas created by a single 200 ps Bessel beam using (a) 200 mJ, (b) 185 mJ, (c) 160 mJ and (d) 133 mJ, 500 ps probe delay, N₂ gas jet](image)

**Figure 4-27**, Interferograms of plasmas created by a single 200 ps Bessel beam using (a) 200 mJ, (b) 185 mJ, (c) 160 mJ and (d) 133 mJ, 500 ps probe delay, N₂ gas jet
Figure 4-28. Diameter of plasma bubble versus pulse energy, 500 ps probe for all points, plasmas are created in N$_2$ jet.

Fitting curve
$D = 13.5 \times (E \times 122)^{0.188}$

$D$ in $\mu m$ and $E$ in mJ

Figure 4-29. (a) Interferogram of the plasma created by a 210 mJ Bessel beam in the N$_2$ jet, the pulse is across the gas jet, 500 ps probe delay; (b) the corresponding modulation diameters and the curve fitting to Gaussian distribution.

Fitting curve
$D = 31.3 \exp\left[-(z-33.47)^2/11000\right]$ 
Both $D$ and $z$ are in $\mu m$. 
However, the origin of the plasma bubbles is still unclear. To fully explain the axial modulation, sophisticated models, either analytical or numerical, are needed. In fact, axial modulation in laser created plasmas is not new and was discussed before. Ping et al successfully explained the periodical structure in plasmas created by nanosecond scale, spherical pulses through a straightforward surface instability model [4.37]. A much more elaborate model of parametric instability, which involved the coupling between the axicon field and a scattered mode of the evolving channel, was introduced by Cooley et al to describe the slow-wave structure they observed in Bessel beam-created plasmas [4.38]. Modification or improvement of these two models might shed more light on our puzzle.

4.5.3 Creation of small diameter plasma waveguides

Although corrugated plasma channels have some particular application in laser based electron accelerators (see, e.g. [4.39]-[4.41]), for X-ray laser experiments we still would like to have uniform plasma waveguides to simplify the dynamics of pulse guiding. We could wait a couple of nanoseconds before injecting the powerful pulse so the plasma bubbles can merge into a single column. However, this column usually has a diameter of \(\sim 50 \mu m\) and would support the propagation of high-order modes, limiting the peak intensity that can be reached by the pulse. Even without a thorough knowledge of the origin of the axial modulation, we still manage to eliminate it experimentally through two methods. One is the modified igniter-heater scheme in which we introduce a low energy femtosecond pulse as the igniter before the arrival of the 200 ps pulse which is now the heater, and both pulses are focused by the axicon lens. The other way is to create the
plasma channels in gas mixtures that have a high concentration of hydrogen. The latter is closely related to soft x-ray lasers since hydrogen can also provide a large amount of cold electrons that are critical for the fast process of the three-body recombination.

Figure 4-30, (a) Interferogram of the plasma created by modified igniter-heater scheme in 400 Torr quiet N$_2$, 30 mJ, 120 fs igniter, 330 mJ, 200 ps heater, 100 ps delay between them, 200 ps probe delay; (b) the corresponding radial density distribution.

In this modified igniter-heater scheme, we expect the femtosecond igniter pulse to generate some free electrons that would seed the collisional ionization of the 200 ps heater pulse. Although the igniter-created plasma is usually weak, sometimes even invisible on the interferograms, its effect on the ionization process of the heater pulse is nonetheless significant as can be seen in Figure 4-30 (a), which shows the plasma created in the chamber filled with 400 Torr nitrogen. The free electrons produced by the igniter dramatically stabilize the interaction between the heater pulse and the gas, practically eliminating most of the axial modulations. In Figure 4-30 (b n in) we plot the radial...
density distribution of this plasma. Very deep density suppression is already established on the axis only 200 ps after the passage of the igniter pulse. The absolute density is also very high, on the order of $10^{20} \text{ cm}^{-3}$.

![Figure 4-31](image)

**Figure 4-31.** Evolution of plasmas created by a single Bessel beam, 200 ps, 230 mJ, in gas jet of 40% $\text{C}_2\text{H}_6$ and 60% $\text{H}_2$, probe delays are (a) 200 ps; (b) 300 ps and (c) 500 ps.

In comparison with the modified igniter-heater scheme, using the gas mixture as the target is more convenient in practice since no extra optics and the consequent alignment are needed. Initially we observed that in pure hydrogen jet, the created plasma channel was always uniform even when we only used a single Bessel beam. This is probably because there is only one electron in each hydrogen atom so its ionization dynamics is much simpler than that of nitrogen or ethane which can generate a variety of ion species when being ionized. Hence, naturally, we expect the doping of hydrogen into the nitrogen or ethane jet would at least alleviate the strong axial modulation. In Figure 4-31, the evolution of the plasmas created in gas jet of 40% ethane and 60% hydrogen are presented. Satisfactory uniformity is achieved along the axis except at the beginning (right end) of the channel where the diameter shrinks a little bit. Figure 4-32 shows the corresponding radial density distributions which maintain a hollow-pipe structure up to
0.5 ns. The axial density suppression flattens out quite quickly but it shouldn’t affect its application in soft x-ray lasers whose powerful pump pulse is only a few microns at focus.

Figure 4-32, Radial density profiles of the plasmas created by a single Bessel beam in the gas mixture (40% C₂H₆ and 60% H₂), 230 mJ pulse energy

Figure 4-33 presents the plasmas generated by the modified igniter-heater scheme in gas mixture of 40% ethane and 60% hydrogen. The uniformity is further improved, with the cone-shaped plasma beginning eliminated. Also, the plasmas are noticeably wider, indicating that more pulse energy is coupled into the plasmas to drive the expansion. In Figure 4-34 we plot the radial plasma densities which are almost the same as with the single Bessel pulse case shown in Figure 4-32. This density clamping implies a possible ionization bottleneck which requires a significant increase of pulse intensity toward the next ionization stage. So the major role of the igniter-created electrons is to help the heater pulse drive the plasma expansion more uniformly and faster.
Chapter 4. Formation of Plasma Waveguides

Figure 4-33, Evolution of plasmas created by modified igniter-heater scheme in gas jet of 40% C$_2$H$_6$ and 60% H$_2$, 66 mJ, 100 fs igniter, 230 mJ, 200 ps heater; probe delays are (a) 200 ps; (b) 300 ps and (c) 500 ps

Figure 4-34, Radial density profiles of the plasmas created by modified igniter-heater scheme (66 mJ igniter, 230 mJ heater) in gas mixture (40% C$_2$H$_6$ and 60% H$_2$), 230 mJ pulse energy

4.6 Conclusion

In this chapter, we presented two types of plasma waveguides not only for soft x-ray lasers but for Raman amplification as well [4.42]. First, we investigated the modified igniter-heater scheme that involved the combination of a 200 ps Bessel beam and a 10 ns
spherical beam. The Bessel beam created a partially ionized, uniform plasma channel that guided the propagation of the spherical beam which effectively deposited its energy into the existing channel for further ionization and expansion drive. Plasma waveguides with 450-500 \( \mu m \) diameters and \( 10^{19} \) cm\(^{-3} \) axial densities were created to guide pulses of large diameters as in the Raman amplification requires. Then we proceeded to explore the generation of small diameter plasma waveguides. In order to eliminate the strong axial modulation associated with the Bessel beam-created plasma channels, we introduced the 100 fs Bessel pulse as the igniter to stabilize the final plasma waveguide. Using the ethane-hydrogen mixture as the gas target also proved to be effective. Both methods, as well as their combination, were used for generating plasma waveguides with 10-30 \( \mu m \) diameter and \( 1-3 \times 10^{19} \) cm\(^{-3} \) on-axis densities, which were within the parameter window for soft x-ray lasers [4.43]. The techniques developed in this investigation will also benefit other applications requiring the guiding of powerful laser pulses.
Chapter 4. Formation of Plasma Waveguides

References:


[4.43] Y. Luo, A. Morozov, S. Suckewer, Suppression of parametric instability in plasmas created by Besselian beams (*in preparation*)
Chapter 5

Spectroscopic Study of Laser-Created Plasmas

Prior to any attempt of direct demonstration of a soft x-ray laser (i.e. gain measurement), it is always instructive and necessary to examine the spectra of the gain medium - plasmas. Such examination can reveal critical information about the plasma conditions, including the dominant ion species, amplitude ratios of the spectral lines, etc, as well as the possibility to achieve gain. In this chapter, we mostly present the spectroscopic study of the ethane plasmas created by 100 fs pulses, which were focused by an off-axis parabolic mirror into intensities near $10^{19}$ W/cm² as required to achieve lasing to ground state at 3.37 nm in CVI ions according to numerical modeling. 25 ps pulses, although too long to pump the lasing on 2-1 transition, were also used to study the effect of pulse duration on the carbon spectra, as well as to explore the conditions to build up the 3-2 population inversion in CVI ions. Other than the pulse duration, two additional factors—the initial gas density and the level of pre-pulse—have also been taken into account. Preliminary results of pulse propagation in plasma waveguides are presented at the end of this chapter.
5.1 Experimental setup

The spectroscopic study of laser-created plasmas, with the experimental setup shown in Figure 5-1, involved two ionization beams and a grazing incidence spectrometer for the VUV and soft x-ray regions. The 100 fs pulse, delivered by the Thales Alpha system, was focused by an off-axis parabolic mirror \((f=150 \text{ mm})\) into the gas jet to create plasmas as the potential lasing medium. The uncompressed, 200 ps pulse from the Positive Light was focused by an axicon to produce a pre-formed plasma waveguide. Radiation from the plasmas traveled through a series of pinholes and slits before being diffracted by a varying line-spacing grating onto a CCD camera that recorded the spectra. A stainless steel tube connected the vacuum chamber, in which plasmas were created, to the spectrometer. Two turbo pumps were employed for pumping the whole vacuum system to a pressure of \(10^{-5}\) to \(10^{-6}\) torr.

Figure 5-1, Experimental setup for the spectroscopic study of laser plasmas
The off-axis parabolic mirror employed in our experiment, manufactured by Janos Technology, had a 60° reflection angle, \textit{i.e.} the angle between the incident and reflected beam. Thus, the 100 fs pulse after parabolic mirror was also at an angle of 60° with respect to the axis of the spectrometer indicated by the horizontal dashed line in Figure 5-1. A 45°-incidence mirror M2 with high reflectivity at 800 nm was used to direct this beam along the axis. The measured reflectivity of M2 under a wide range of incident angles from 30° to 80° was between 60% and near 100%, resulting in > 75% overall reflectivity for the focusing beam on M2.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure5-2.png}
\caption{CCD image of the 100 fs pulse at the focus of the off-axis parabolic mirror}
\end{figure}

One of the key elements in this experiment was the beam profile at the focus. Near diffraction-limited focus of the pulse is desirable in achieving the highest possible intensity (over $5 \times 10^{18} \text{ W/cm}^2$) in order to create totally stripped plasmas. The way we monitored the focal profile of the 100 fs pulse was to insert a mirror for 1 \textmu m beams before the gas jet. About 4% of the 800 nm pulse was reflected out and picked up by a CCD camera after passing through an objective lens. The surface of the mirror for 1 \textmu m beams was of good optical quality. Therefore, no phase distortion would be introduced into the pulse upon the reflection, resulting in reliable monitoring of the beam profile.
Figure 5-2 shows the original CCD image of the 100 fs pulse at the focus. Despite some structures at the edge of the beam, the central bright spot is smooth and symmetric.

Figure 5-3, (a) Computer reconstructed image of the 100 fs pulse at the focus of the off-axis parabolic mirror and (b) its radial intensity distribution with 300 mJ input energy.

To quantitatively investigate the focal profile, we reconstructed the CCD image in MATLAB assuming 300 mJ for the 100 fs pulse. Figure 5-3 (a) clearly illustrates the details of the edge structure. It can be seen that most of the pulse energy concentrates in the symmetric central spot while the peripheral wings are almost one order of magnitude less intense than the center. In Figure 5-3 (b) we plot the radial intensity of the pulse at the focus for 300 mJ pulse energy. Two features are worth to mention here. First, the FWHM of the beam is <3.6 \( \mu m \), which is about 1.5 times diffraction limit. Here, the beam diameter is 60 mm and the focal length of the parabolic mirror is 150 mm. Second, the average intensity within the FWHM approaches \( 10^{19} \) W/cm\(^2\), which should be sufficiently high for the production of abundant CVI ions from the C\(_2\)H\(_6\) gas target. The slight asymmetry of the radial profile on the right might come from the varying reflectivity of M2 as mentioned before. But it is quite minor comparing with the overall quality of the beam.
The assembly of the axicon was more complicated as seen from the inset in Figure 5-1. The 200 ps pulse was focused by an axicon whose axis is 45° with respect to the horizontal direction. Then a mirror M3, with a conical hole in the center to pass through the 100 fs pulse, reflected the outer part of the Bessel pulse to form a line focus above the gas jet. Only the rays that contributed to the line focus were shown in the figure. We didn’t shoot the 100 fs pulse directly through the hole of the axicon, as in Chapter 4, because the F number of the parabolic mirror was so small (F/#=2.5) that the resulted half angle (11.3°) of its focal zone was larger than that of the axicon (9.73°). Therefore, the 100 fs would inevitably clip on the axicon and cause damages if we wanted to focus the 100 fs pulse at the entrance of the plasma waveguides, which was usually the case for pulse guiding. Using the holed mirror M3, in fact, didn’t solve the clipping problem, but mirrors were much less expensive than the axicon and could be easily replaced.

The homemade soft x-ray spectrometer used a Hamamatsu variable line-spacing grating [5.1] as the diffracting element so the whole system was compact and almost astigmatism-free. The grating had a characteristic line spacing of 1200 lines/mm which gave a resolution power close to $10^5$ with the illumination width of 8-10 cm. A 1024×1024 PIXIS CCD camera from Princeton Instrument then picked up the dispersed soft x-ray signal from the plasmas. The CCD detector was mounted on a horizontal sliding rail that was perpendicular to the axis of the spectrometer. Therefore, by moving the detector from one end to the other, we could record radiations in the spectral region from 1.8 nm up to 26 nm. The calibration of the spectrometer, as well as the subsequent processing of the spectra, was mostly conducted in the WinSpec software that came with the camera.
5.2 Calibration of the spectrometer

Before applying the spectrometer into real experiments, we calibrated its wavelength using the boron plasma as the radiation source. As shown in Figure 5-4, we focused a 10 ns pulse of 30-50 mJ, delivered by our NG 24 Nd: YAG laser system, through a spherical lens of 15 cm focal length onto the sharp edge of a solid boron piece to create the plasma. The boron target was fixed on a holder that could slide perpendicularly to the axis of the spectrometer. By scanning the position of the target, we could optimize the signal on the CCD camera, and at the same time the tip of the target would accurately mark the position of the spectrometer axis.

![Figure 5-4. Schematic of the spectrometer calibration setup](image)

Figure 5-5 (a) presents the original spectra of boron plasmas on the CCD across its full movement range from 14 mm to 40 mm. The step size was chosen as 5-6 mm so we could always have several overlapping lines at two successive positions and thus calibrate any new CCD position using the line identification from the previous step. Also, these spectra illustrate one advantage of boron plasmas as the source. They have sufficient number of lines to achieve a reliable calibration over the entire CCD plane through linear or polynomial interpolation. But the number of lines is not yet too large to impose undue difficulty for line identification.
Chapter 5. Spectroscopic Study of Laser Created Plasmas

Figure 5-5, (a) Original spectra of Boron plasma created by 10 ns pulses at variable CCD positions from 14 mm to 40 mm, and (b) its calibrated spectral profile with peaks numbered from 1 to 19. Notice that some of the strong lines are over-exposed in order to show the weaker lines.

Figure 5-5 (b) shows the merged spectra of the boron plasmas with peaks numbered from 1 to 19. It is not difficult to see that these peaks are clustered: every five peaks (except the last four) form a group and the total 19 peaks can be divided into four groups. Table 5-1 summarizes the identification of the first five peaks, or the first group. It turns out that all the lines are from BIV ions (He-like boron), with the ground state as their common lower level of the transitions. The upper levels range from 1s6p to 1s2p, resulting in emission wavelength from 4.89 nm up to 6.03 nm. A careful calibration also confirms our observation of the peak clusters. The second to fourth groups are exactly the second- to fourth-order diffraction of the first group. The fourth order of the 1s5p-1s2
transition is barely seen in the spectra because the line intensities drop by nearly 85% as the diffraction order increases from one to four. The 1s5p-1s² transition, which has only 20 counts at the first order, has only 3 counts at the fourth order, comparable to the noise level. This near-complete list of the observed BIV radiation in the spectral region of our primary interest also reinforces the reliability of the calibration. With this confidence in mind, we can now proceed to analyze unknown spectra from the spectrometer.

### Table 5-1, Calibration of the boron plasma emission spectra

<table>
<thead>
<tr>
<th>Peak number</th>
<th>Wavelength (nm)</th>
<th>Ion</th>
<th>Transition</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>4.89</td>
<td>BIV</td>
<td>1s6p-1s²</td>
</tr>
<tr>
<td>2</td>
<td>4.95</td>
<td>BIV</td>
<td>1s5p-1s²</td>
</tr>
<tr>
<td>3</td>
<td>5.04</td>
<td>BIV</td>
<td>1s4p-1s²</td>
</tr>
<tr>
<td>4</td>
<td>5.27</td>
<td>BIV</td>
<td>1s3p-1s²</td>
</tr>
<tr>
<td>5</td>
<td>6.03</td>
<td>BIV</td>
<td>1s2p-1s²</td>
</tr>
</tbody>
</table>

5.3 Contrast improvement using saturable absorbers

In experiments of high-intensity laser-matter or laser-plasma interactions, the contrast ratio of the laser, defined as the ratio between laser peak intensity and any pre-pulse or pedestal intensity, usually has to be taken into account and improved if necessary. This is particularly true in experiments involving charged-particle acceleration or the formation of plasma waveguides, since large pre-pulse or pedestal tends to create pre-plasma, which might significantly alter the dynamics of energy coupling between subsequent main pulse and the target, or simply disrupt the propagation of the main pulse.
For high power laser systems based on the CPA technique, there are three major sources of pre-pulse, each of which corresponds to a characteristic time scale. The first source is from the limited extinction ratio of the pulse slicer after the oscillator or regenerative amplifier. In ideal case, only one pulse can pass through the slicer in each of its working cycle and enter the subsequent amplifiers. However, the limited extinction ratio usually allows the passage of part of the neighboring pulses which will undergo similar amplification and evolve into large pre-pulses. They are usually on nanosecond scale, from a few ns to 20 ns. The second source is the amplified spontaneous emission (ASE) in the amplifiers. The spontaneous fluorescence or sometimes even noise in the pumped laser crystals gets amplified and propagates together with the main pulse. ASE usually results in a long pedestal both before and after the main pulse, and it is typical in the range of several ns long, determined by the spontaneous emission rate. The third possible source originates from the grating compressor. The imperfect compensation of the chirp may result in a pre-pulse of tens of picoseconds. In the Thales Alpha laser system, the first source makes an important contribution to the pre-pulse. We observed a clear pre-plasma (see Figure 5-7 (a)) about 2 ns before the main pulse. The pre-plasma expands to almost 100 \( \mu m \) wide by the time the main pulse arrives. It is also sensitive to the timing of the Pockel’s cell in the pulse slicer, implying at least part of its origin.

Several methods have been developed to improve the laser contrast, or to manipulate the pre-pulse and pedestal to minimize its negative effects in experiments. This includes saturable absorbers [5.2]-[5.3], thin carbon foil [5.4], double CPA system [5.5], nonlinear birefringent crystals [5.6], cross-polarized wave generation [5.7], plasma mirrors [5.8]-[5.9] and second harmonic generation [5.10]. We choose to use saturable absorbers
because they can be conveniently integrated into or removed from our laser system depending on practical requirements. The special feature of saturable absorbers that is pertinent to laser contrast improvement is its nonlinear absorption with respect to incident intensity. To be specific, they absorb most of the energy of low intensity pulses but become nearly transparent if the incident intensity is above a certain threshold. After the pulse slicer following the regenerative amplifier, the laser beam in fact contain several pulses, including a main pulse that is intended to be amplified, and a few neighboring pulses that leak through the slicer but they are much less intense. Therefore, if the saturable absorber is properly chosen, the main pulse would largely pass through but its neighbors would be mostly absorbed, resulting in a larger intensity ratio, \textit{i.e.} larger contrast, between the main pulse and the neighboring ones.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{saturable_absorber_transmission_curve.png}
\caption{Transmission curve of the RG 850 saturable absorbers with 1 mm and 1.5 mm thickness}
\end{figure}
The saturable absorber we selected was the Thorlab RG850 colored-glass filter made from Schott glasses. Two absorbers with 1 mm and 1.5 mm thickness have been used and their transmission curves under different incident intensities were characterized before being integrated into the laser system. We focused the laser pulse from the regenerative amplifier through a spherical lens of 1 m focal length and installed the absorbers on a rail along the beam path after the lens. By simply moving the absorber along the rail to different distances behind the lens, we were able to acquire a wide range of incident intensities and measure their transmission. The dynamic range of incident intensity was further extended (toward lower intensity) by placing neutral-density (ND) filters before the lens. The pulse immediately after the regenerative amplifier was 200 ps, 1.3 mJ and 1 cm wide. The incident intensity on the absorbers could thus be estimated using a beam size calculated from geometrical optics considering the absorber-lens distance. The transmitted energy was measured by a Gentech-EO power meter, which had a resolution of a few $\mu$J, comparable to the noise level of the system.

In Figure 5-6 we present the transmission of the RG 850 saturable absorbers as a function of incident intensity. Both absorbers show clear near-saturation behavior at the two ends of the intensity range. At the low intensity end, the 1 mm absorber transmits roughly 20% of the pulse, much higher than the 6-7% transmission of the 1.5 mm piece. The distinction is much smaller at the high intensity end, where the transmission only slightly differs (75% for 1 mm and 70% for 1.5 mm). The largest contrast enhancement occurs when the transmission difference between the main pulse and the pre-pulse is also maximized, i.e. the main pulse is within the high-intensity saturation end while its
neighbors are within the low-intensity end. In real experiments, the situation might not be so ideal but significant improvement is still promising.

We then placed the RG850 absorbers before the two-stage final amplifier of the Thales Alpha system to investigate their effects on the laser beam contrast. This location was chosen for practical reasons. The absorbers might not be exactly perpendicular to the incident pulse so it could shift the beam position. Also, instead of plates of perfectly parallel surfaces, the absorbers might be slightly wedged and thus altered the direction of the beam as well. The difficulty of correcting this absorber-induced misalignment was minimized when the absorbers were very close to the two four-pass amplifiers.

![Figure 5-7](image_url)

**Figure 5-7**, Shadowgrams of the pre-plasmas created by 100 fs pulse, the probe is right before the arrival of the 100 fs (a) without any saturable absorber and (b) with both RG 850 filters before the final amplifier of Thales Alpha system, the dramatically depressed pre-plasma is within the dashed line rectangle

Usually the laser pulse contrast ratio is quantitatively measured by a third-order cross-correlator [5.2]. However, since we were most concerned with the pre-plasmas that might interrupt the pulse propagation, it was thus convenient and straightforward for us to check the contrast improvement by directly monitoring the pre-plasmas through
shadowgrams. The best result was achieved when both RG850 absorbers were used (equivalent to a 2.5 mm absorption length) and the comparison of the pre-plasmas is shown in Figure 5-7. Without the absorbers, the pre-plasma appears approximately 2 ns before the main pulse, and it evolves to about 300 $\mu m$ long and 100 $\mu m$ wide upon the arrival of the main pulse. The insertion of the two absorbers not only shrinks the size of the pre-plasmas but decreases its density as well, indicated by the much weaker front of the shock wave. It is worth mentioning that the role of the pre-plasma in achieving the final gain of soft x-ray laser is still debatable (e.g. see [5.4]). Nevertheless, it is always helpful to have some control over the pre-plasmas so we can have more options in optimizing the performance of the soft x-ray laser. We have also compared the final output energy of the laser with and without the absorbers, and it turned out that less than 25% energy was lost. Although each RG850 filter absorbs more than 25% energy of the incident pulse, the amplifiers are operating in a saturated amplification regime so part of the loss on the filter is compensated by subsequent amplification.

5.4 Emission spectra of ethane plasmas

5.4.1 Acquisition of ethane plasma spectra

The longitudinal configuration (laser pulses are parallel to the spectrometer axis) of the laser-gas interaction imposed three unique challenges that didn’t exist during the calibration, in which the laser pulse was nearly perpendicular to the solid surface where the plasma was created. These challenges had to be addressed before we were able to acquire consistent and reliable spectra of the plasmas in ethane jet.
The first challenge was to suppress on the CCD camera the 800 nm radiation from the ionizing pulse. Since the laser pulse was propagating toward the spectrometer along its axis, not only did a small portion of the beam follow the same path as the soft x-ray signal, but another portion, scattered by the plasma into every direction, also underwent multiple reflections inside the vacuum chamber and the spectrometer. Both of these two sources of 800 nm radiation would find their way onto the CCD and introduce unwanted noises over the spectra or even render the spectra irresolvable in serious cases. For each source, we took corresponding measures. An aluminum filter of 30 nm thickness was placed in front of the grating to block the 800 nm radiation along the spectrometer axis, while transmitting most of the soft x-ray signal [5.11]. As for the scattered portion, we put black papers of low reflectivity on as many reflection surfaces as possible. The noise could not be totally eliminated, but it was reduced to an acceptable level.

The second challenge was the accurate repositioning of the plasmas on a daily basis. In the calibration, the plasma always formed on the boron target’s tip, whose position, once found to be correct, stayed there. However, for the laser-gas interaction, a small misalignment of the laser system could easily shift the plasmas out of the spectrometer’s field of view. To solve this problem, we first used a LiF target to find out the spectrometer axis and marked it by a He-Ne beam. Then we placed a 200 $\mu$m copper pinhole, which was installed on a kinematic mount, right above the gas jet and aligned it to transmit the He-Ne beam. Therefore, whenever uncertainty arised about the alignment, we could always put back the pinhole to recover the correct plasma position on the spectrometer axis. The 200 $\mu$m aperture proved to be accurate enough considering the 1 mm field of view of our spectrometer.
The third challenge was the strong absorption of the soft x-rays in the gas jet. For example, the 1/e absorption depth for 100 eV (12.4 nm) and 1 keV (1.24 nm) photons in N\textsubscript{2} gas of 1 atm is only 190 nm and 9 \( \mu \text{m} \) respectively [5.12]. As a result, the plasmas had to cover the entire gas jet in order for sufficient soft x-ray signals to reach the detector. For the pulse with a Rayleigh length of only 50 \( \mu \text{m} \), propagating over the entire gas jet of 5 mm—100 Rayleigh lengths—is extremely demanding. At this stage of experiment when the main goal was to examine the plasma conditions, we rotated the gas jet by 90\(^\circ\) so the effective gas length became 0.5 mm, significantly reducing the absorption. Therefore, the spectra presented in this chapter were obtained using a single 100 fs or 25 ps pulses without concerning the creation of plasma channels. However, for the gain measurement, it is advantageous and necessary to have the ability to extend the length of the lasing medium via the plasma waveguide method. At the end of this chapter, we present the pulse propagation in the plasma waveguides and the possibility of extending the favorable plasma conditions to a much longer distance.

\textbf{Figure 5-8,} A sample spectrum of ethane plasma on the CCD camera; the plasma is created by a 25 ps, 200 mJ laser pulse, and the spectral range is from 2.8 nm (left end) to 8.6 nm (right end)
Figure 5-8 presents a typical spectrum of ethane plasma created by a 25 ps pulse in the 2.8 nm-8.6 nm range. Despite the background noise, the spectra lines are still clearly visible. The whole system was also robust to minor misalignment of the laser and our daily routine correction, so the data collected at different days are consistent and can be compared with confidence. In our experiment, three parameters—duration of the ionizing pulse, initial gas density and level of pre-pulse—could be conveniently adjusted to optimize the final spectra, as well as to investigate the underlying physics. The following sections present how the carbon spectra responded to the change of these factors.

5.4.2 Pulse duration effect on carbon spectra

Our first set of spectra was obtained in the ethane gas jet using pulses of different durations, from 100 fs up to 25 ps, for ionization. One of the two gratings in the final grating compressor of the Thales system was mounted on a rail that was parallel to the direction of the pulse. By sliding the grating we were able to compensate the chirp to different degrees and thus adjust the pulse duration of the output. The grating pair and the rail were precisely aligned such that the adjustment of pulse duration would not introduce any misalignment to the rest of the system. Pulse duration at the two temporal ends (100 fs and 25 ps) was accurately measured using the auto-correlator, while the intermediate ones (1 ps, 5 ps and 10 ps) were estimated from the linear interpolation. The shortest pulse duration of 100 fs does not correspond to the end of the rail. That is because the shortest duration is achieved by a near-exact compensation of the chirp
applied by the stretcher: over-compensation is as bad as under-compensation in preventing the pulse from reaching the shortest duration.

Figure 5-9, Carbon spectra of ethane plasmas created by different pulse duration, 200 mJ pulse energy for all the cases

Figure 5-9 shows the CV and CVI spectra in 2.8 – 8.7 nm range, obtained from plasmas created in ethane jet by pulses of different duration (100 fs, 1 ps, 5 ps, 10 ps and 25 ps) but with the same pulse energy of 200 mJ. The spectral lines of CVI ions (hydrogen-like) are identified by bold letters and CV ions (helium-like) in regular font. Most of the CVI lines, except the line from 2-1 transition at 3.37 nm, are from the 2nd order diffraction due to the limited operating range of the detector and the high-level background signal from the zero-order diffraction. In fact, in our experiment it is usually more convenient and more precise to study the 2nd order diffraction of the emission lines
below 3 nm, since the CCD detector has a higher sensitivity at larger diffraction angles, corresponding to higher orders of diffraction. Also, the noise level at the left end of the CCD camera (see e.g. Figure 5-8) is higher due to the closer distance to the zero-order diffraction (secular reflection).

Since one of the critical elements for any soft x-ray laser to work is to create abundant lasant ions—CVI ions in our case—it is thus instructive to study what kind of ions can be produced under the irradiation of pulses with different durations. It is clear that in all cases presented in Figure 5-9, CVI ions dominate in the plasmas, evidenced by significantly stronger lines of CVI ions than those of CV ions for the same type of transitions to ground states \( n \rightarrow 1 \), where \( n \) is the principal quantum number in hydrogen-like CVI and helium-like CV ions. For example, the line from the \( 1s2p-1s^2 \) transition at 4.02 nm, which has the highest spontaneous emission coefficient in CV ions, is almost negligible in comparison with CVI lines in most of the experiments.

Although both the 100 fs and 25 fs pulse are able to create CVI-dominated plasmas, the underlying ionization mechanism is very different. The ultra-short and ultra-intense 100 fs pulse strips the electrons from their parent atoms or ions mostly through Optical Field Induced (OFI) ionization, while the 25 ps pulse leaves enough time for avalanche ionization to establish its dominance. OFI plasma is advantageous for soft x-ray lasers, especially for lasing to ground states, due to its highly non-Maxwellian electron temperature profile with excess electrons in the low energy region, which is very important for a fast process of three-body recombination. Moreover, ultrashort pulse duration is also required in order to deliver the pump energy within a time scale shorter than the upper level radiative lifetime (e.g. 1.6 ps for \( n=2 \) of CVI ions). Nevertheless, as
to lasing at the 3-2 transition, 25 ps or even longer pulses could still be useful, provided that sufficiently fast cooling is available [5.13]-[5.14].

Besides the abundance of CVI ions, another striking feature of this set of spectra is the indication of population inversion between 2\textsuperscript{nd} and 3\textsuperscript{rd} excited level in CVI. In plasmas of local thermodynamic equilibrium (LTE), estimation using Saha equation gives that the line intensity of 3-1 transition versus 2-1 transition should not exceed 1:3 for an electron density of 10\textsuperscript{19} cm\textsuperscript{-3} [5.15]. But in our spectra, this line intensity ratio is almost 1:1, strongly indicating the 3-2 population inversion. This implies the dominance of three-body recombination and the populating of lower levels through cascade collisional de-excitation. If the reverse would be true, \textit{i.e.} the photon-recombination outruns the three-body recombination, lower levels would be quickly populated so there would be no chance for any 3-2 population inversion to be established. One underlying condition of three-body recombination’s dominance is a relatively low plasma temperature. In this experiment, we use pure ethane as the gas target. If we add some ‘buffer gas’ such as hydrogen that has a much lower ionization potential (13.6 eV) than that of CV (392 eV) or CVI (490 eV), the plasma temperature would be further reduced according to numerical calculations, so the plasma conditions would be much more favorable for lasing to ground states. However, we cannot infer in any simple way the population inversion between n=2 and the ground state from this spectra, because there are no spontaneous emission lines in this spectra from the ground state. The way to verify the 2-1 population inversion is by gain measurement. Nevertheless the low temperature of the plasma and the resulting fast recombination to excited states of CVI and CV ions makes the possibility of lasing to ground states in both of these ions very promising.
5.4.3 Gas density effect and line broadening

Gas density plays a complicated role in soft x-ray lasers. It not only determines the plasma density and thus the emission intensity, but also influences the dynamics of laser-gas interaction and the spectral width. The tuning of initial gas density in this experiment was realized by adjusting the jet opening time with respect to the arrival of the laser pulse through a digital signal-generator from Stanford Research Systems (SRS). Laser pulses of 25 ps and 100 fs were used to investigate the plasma conditions for lasing at 3-2 and 2-1 transitions respectively.

Out of the roughly 1 ms duration of the jet opening for each shot, we only operated within one small window (less than 0.1 ms time span) in the leading edge. There existed an optimal gas density for spectra acquisition. A density that was too high aggravated the already severe nonlinear effects that might disrupt the pulse propagation, so the plasmas could not span the entire gas jet, preventing the signals from reaching the CCD. This was verified by the shadowgraphic study of the plasmas: spectra could only be observed when the plasmas extended beyond the edge of the gas jet. A density that was too low was equally detrimental in that the emitted signal was too weak to rise above the background noise. These two factors constrained the < 0.1 ms timing window (corresponding to about 100% density change, see Figure 3-9) within which we could obtain reliable spectra.

The reason to work at the leading edge of the jet opening, instead of the trailing one that provides comparable density, is to minimize the spread of gases when the laser pulse arrived. The lateral spreading would directly reduce the density gradient along the axis, forcing the pulse to interact with the low-density region in which the nonlinear
instabilities might disturb the focus of the pulse and prevent it from reaching the peak intensity, or simply disrupt the propagation (e.g. formation of multiple filaments). For the same reason, the laser pulse has to be within 0.6 mm above the surface of the nozzle: the gas density gradient drops at higher positions (see Figure 3-10). Moreover, working at the trailing edge of the jet opening would allow the gases a sufficient time to diffuse into the chamber and increase the background pressure. These prevailing gases, although with modest densities comparing to the jet, would still strongly absorb the soft x-ray signal and impose undue difficulty for data acquisition.

![Figure 5-10](image-url)  

**Figure 5-10.** Carbon spectra of ethane plasmas created by 25 ps, 200 mJ pulse for different gas densities

In Figure 5-10 and Figure 5-11 we present the CV and CVI spectra from ethane plasmas created by 25 ps and 100 fs pulses, respectively. For each case, various initial gas densities have been used. As the jet opening time increases from 200.12 ms to
200.20ms, the corresponding initial gas density decreases roughly by 50%. The timing window, within which we could obtain reproducible spectra, has been observed to be closely related to the pulse duration. To be specific, the 100 fs pulse has a much narrower timing window (0.04 ms as found from experiment) than that of 25 ps (>0.08 ms), a result of the more severe nonlinearity in its interaction with the gas. Similar to Figure 5-9, the decrease of pulse duration, from 25 ps to 100 fs, also improved the dominance of CVI lines in the spectra. The 2\textsuperscript{nd} order of 1s2p-1s\textsuperscript{2} transition line of CV ions is noticeably weaker for the spectra using 100 fs pulses.

![Figure 5-11](image)

**Figure 5-11, Carbon spectra of ethane plasmas created by 100 fs, 200 mJ pulse for different gas densities**

The density effect on both spectra is firstly reflected by the relative intensity between the 3-1 and 2-1 transition lines. As the gas density goes up (from the top to the bottom in the figure), the intensity ratio of the 3-1 to 2-1 transition lines also increases. For 25 ps
pulses, this ratio increases from 1:1.1 to 1.5:1, producing a 65% enhancement. As for 100 fs pulses, this enhancement is approaching 90%, from 1.5:1 to 1.25:1, despite that a narrower timing window has been used. This increase of the relative population in level \( n=3 \) with respect to level \( n=2 \) can be ascribed to the enhanced rate of collisional de-excitation, which has a rate that is proportional to the electron density. Higher gas density produces higher electron density. The correspondingly enhanced collisional de-excitation dominates over the radiative de-excitation and transfers more electrons to higher energy levels (see Chapter 1, section 1.2.2). But such enhancement is limited by the disruption of pulse propagation due to nonlinearity and instabilities as discussed earlier in this section.

From Figure 5-10 and Figure 5-11, we can also obtain some information about the broadening of the spectral lines. Spectral broadening is closely related to the gain cross section \( \sigma_{\text{gain}} \) according to \( \sigma_{\text{gain}} = \left( \frac{\lambda^2}{8\pi c\Delta\lambda} \right) A_{ul} \), where \( \lambda \) is the wavelength, \( \Delta\lambda \) is the spectral width of the lasing transition line, and \( A_{ul} \) is the spontaneous transition probability (Einstein A coefficient) between the upper and lower levels that are involved in lasing. Therefore, a narrow line of lasing emission is important to achieve maximum gain. In our experiment, although the instantaneous electric field during the laser-plasma interaction is very high (\( >10^{11} \) V/cm), the acquired spectra are mostly collected long after the passage of the pulse, so ac Stark broadening can be safely assumed negligible. Pressure broadening is also playing a minor role. A careful measurement reveals that the line width of each peak, in general, decreases along with the density. For example, in Figure 5-10, the FWHM of the 2\textsuperscript{nd} order of 5-1 to 2-1 transitions of CVI ions decreases from 0.22-0.23 Å at 200.12 ms to 0.17-0.19 Å at 200.20 ms. A similar trend, but with
slightly smaller amount of narrowing is also found in Figure 5-11, which presents the data with 100 fs pulses and a narrower timing window for jet opening time (smaller density change). The decrease of spectral width is comparable to the resolution of the detector, which is approximately 0.05 Å per pixel size. The dominant contribution to the observed line width comes from the instrumental broadening, which is about 120 mÅ and orders of magnitudes larger than the natural line width. Therefore, we ruled out the possibility of collisional or Stark broadening in causing this change of line width. Instead, we believe that the change of line width might be attributed to the dielectronic satellite lines originating from $2l/2l'$ or $2l3l'$ levels of CV ions [5.16]. Take the Lyman-α line of CVI ions for example. Some of these satellite lines are so close to the Lyman-α line that they cannot be resolved by our spectrometer. The observed Lyman-α line is in fact the superposition of several lines. The upper levels of these satellite transitions are populated either by dielectronic recombination or electron collisional excitation, both of which are directly a function of the electron density. As a result, the decrease of initial gas density leads to the decrease of satellite line intensity, thus reducing the observed line width.

5.4.4 Pre-pulse effect on the spectra

The debate over the role played by the pre-pulse in spectra can usually be reduced to the debate over pre-plasmas. On one hand, pre-plasmas can provide seed electrons for further ionization. So they might boost the coupling of laser energy into the plasma and improve the absorption, particularly for pulses of longer duration that strip electrons through collisions. On the other hand, however, pre-plasmas might also disturb the propagation of pulses. The shock wave front (i.e. the boundary) of the pre-plasmas has
an extremely sharp density gradient which is capable of refracting and scattering part of the pulses that are incident upon it. This might induce an energy loss from the pulse, or prevent the pulse from reaching its ideal focus and thus peak intensity. Due to the dual role of the pre-plasma, we performed experiments with two different levels of pre-plasmas while controlling the other parameters as best as possible, in order to examine the pre-pulse effect on the spectra directly. The way to suppress the pre-plasma was to insert the two pieces of RG850 saturable absorbers as introduced in section 5.3.

![Image of carbon spectra](image)

**Figure 5-12**, Carbon spectra of ethane plasmas created by 25 ps pulses with and without the saturable absorber RG850, 200 mJ pulse energy

The effect of pre-plasma on the carbon spectra using 25 ps pulse is shown in Figure 5-12 where ‘RG850 on’ means that the pre-pulse is largely suppressed. The large difference in the amplitudes of the peaks between the two set of spectra is a result of different number of shots used in obtaining the spectra. 30 shots were used for 25 ps...
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pulses while only 12 for 100 fs pulses. Since our spectrometer has not been calibrated in absolute intensity, it is more informative to examine the relative intensity within each spectrum. The ratio between 3-1 and 2-1 emission line, and the possible 3-2 population inversion, stays almost the same as the pre-pulse is suppressed. However, 1s2p-1s2 transition at 4.02 nm, as well as its 2nd order at 8.04 nm, shrinks together with the pre-plasma. Therefore, suppression of the pre-pulse is constructive, at least in improving the dominance of the CVI lines.

Figure 5-13, Carbon spectra of ethane plasmas created by 100 fs pulses with and without the saturable absorber RG850, 200 mJ pulse energy

Figure 5-13 presents the CVI and CV spectra with and without saturable absorbers using the 100 fs pulse with 200 mJ pulse energy for all cases. In this set of data, the spectral range was extended on the shorter end to 2.5 nm and thus included the 3-1 transition line of CVI ions at 2.84 nm, at the sacrifice of the 2nd order of 1s2p-1s2...
transition line of CV ions. In both spectra, with and without the saturable absorbers, we observe indication of 3-2 population inversion directly from the 1\textsuperscript{st} order peaks of 3-1 and 2-1 transition lines at 2.84 nm and 3.37 nm, respectively. But the spectra with the pre-pulse suppressed show several features that are particularly encouraging. The first feature is the noticeably enhanced ratio of line intensities between 3-1 and 2-1 transition (2\textsuperscript{nd} order peaks), from 1:1.25 to 1.25:1, increasing by more than 50%.

The second feature is more subtle but more important. With the pre-pulse suppressed, we see that the 2\textsuperscript{nd} order of 1s3p-1s\textsuperscript{2} transition line increases. This reveals the potential to achieve lasing to the ground state in CV ions after the completion of lasing at the 2-1 transition in CVI ions—practically a cascade lasing. If we have lasing at 2-1 transition in CVI ions, the exponential amplification of stimulation emission quickly populates the ground level. This process is so fast that the electrons produced from tunneling ionization don’t even have sufficient time to complete the Maxwellization. Therefore, these non-Maxwellian electrons, most of which are in the low energy region, recombine with the abundant CVI ions at ground states through three-body recombination, initiating a similar process of recombination soft X-ray lasing in CV ions. Moreover, the faster transfer to ground states in CVI ions also leaves the plasma channel less time for expansion, preserving a higher plasma density when CV ions are created. Both of these two factors lead to stronger emission lines from CV ions, as we observed in Figure 5-13. If the electrons are already in equilibrium when CVI ions are recombing into CV ions, the ground states of CV ions would be predominately populated and it would be much less likely to observe any strong emission lines from CV ions. At this stage, due to limited diagnostics, we are not ruling out other possibilities that may cause the increase of CV
lines when the pre-pulse is suppressed. For example, the 100 fs pulse with suppressed pre-pulse may create higher plasma density and accelerate the population to the ground level of CVI ions through collisional de-excitation.

![Figure 5-14](image)

**Figure 5-14.** Carbon spectra of ethane plasmas created by 100 fs pulse with suppressed pre-pulse for various initial gas densities, 200 mJ pulse energy

The third feature is the comparable intensities of 5-1, 4-1 and 2-1 transition lines, indicating the possible 5-2 and 4-2 population inversion if we compare it with the Saha-LTE estimation [5.15]. We also find similar indication for the 5-4 population inversion. To verify this observation, we monitored the carbon spectra using 100 fs pulses with suppressed pre-pulse for different initial gas densities, with the results presented in Figure 5-14. Although the allowable timing window further decreased to about 0.02 ms, the variation of initial gas density and the subsequent variation of electron density resulted in
higher intensity of 5-1 line than that of 4-1 line for the jet opening time at 200.22 ms, which reinforced the indication of possible 5-4 population inversion.

This feature is in similarity to the spectra obtained by Krushelnick et al. in our laboratory [5.4] when conditions of the lasing to the ground state of hydrogen-like LiIII ions at 13.5 nm were examined. At that time, it was also possible to change the relative populations of the upper levels (n=3, 4, 5) of the emission lines by adjusting pumping pulse energies and the plasma densities. Later, when extension of the plasma channel was made possible with the help of a microcapillary, gain as high as 11 cm⁻¹ was measured with a maximum gain-length product GL in single shots to be ~6.5. This comparison study makes good prospect of lasing to ground states in CVI at 3.37 nm through the adjustment of pumping pulse energy and plasma densities. Limited by the performance of the laser system and losses along the beam path, only 200 mJ is available when the pulse arrives at the gas jet. With the upgrade of the laser performance which is in our schedule, we would have more flexibility and thus more confidence in the achievement of high gain.

5.5 Preliminary results of pulse propagation in plasma waveguide

The condition for gain measurement in XUV and X-ray region is to have extended plasmas with large population inversion such that during a single pass the amplified lasing line becomes dominant in the spectra, much stronger than the spontaneous emission from the plasma channel. We employed the plasma waveguide method, which was described in Chapter 4, to prevent the radiation spreading induced by refraction, scattering and a variety of instabilities. The plasma waveguides, pre-formed by a Bessel
beam of relatively low intensity, are partially-ionized, with usually only one or two electrons stripped from each atom. Thus, additional ionization is inevitable when the powerful beam propagates in the waveguide, aggravating the already complicated picture of the pulse response to different plasma density profiles. Lacking an elaborate simulation, we proceeded to investigate this issue experimentally.

Figure 5-15. Demonstration of the radiation confinement in pre-formed plasma waveguides, shadowgrams of (a) plasma channel created by 190 mJ, 200 ps Bessel beams only; (b) plasma created by 100 mJ, 25 ps spherical beam focused by parabolic mirror and (c) the propagation of 25 ps pulse in the pre-formed plasma waveguide, 2 ns between the two pulses

The plasma waveguides were created by the 200 ps Bessel beam, delivered by Positive Light in this experiment. We first attempted to guide the 25 ps pulse, instead of the 100 fs one, for two reasons. First there was less severity of instabilities associated with the propagation of 25 ps pulses. Second it was experimentally easier to obtain reproducible spectra using 25 ps pulses. It would be very informative to examine how the pulse guiding influences the spectra. The two pulses—the Bessel pulse and the 25 ps pulse—were synchronized with an optical delay line to adjust the relative timing between the two beams. A 400 nm, 200 fs pulse, frequency-doubled from the compressed output beam of Positive Light, was used as the probe beam to take shadowgrams of the plasmas. Both ionization pulses propagated near-perpendicularly to the orifice of the gas jet. The
effective length of the gas target was adjusted by changing the distance between pulses and the surface of the nozzle (see section 3.3.1 in Chapter 3).

Figure 5-15 presents the preliminary results of how the 25 ps pulse propagates in the pre-formed plasma waveguides and boosts the plasma density through additional ionization. With 190 mJ of energy, capped by the performance of the laser, the Bessel beam can only produce a short plasma waveguide of 0.3 mm. Also, the single 100 mJ, 25 ps pulse stops propagating even before reaching the center of the gas jet (the bright area in the shadowgram). But the combination of these two pulses is definitely constructive. The 25 ps pulse is well confined within the plasma waveguide after its entrance, and it propagates along the entire length of the waveguide while inducing additional ionization, evidenced by the larger diameter and darker hue of its shadow.

Based on this encouraging preliminary result, an urgent work at this stage is to improve the performance of the Positive Light laser system so that more energy can be available for the Bessel beam in order to create a high quality plasma waveguide. Here we don’t split energy from the Thales System, as we did in Chapter 4, because it is critical to concentrate as much energy as possible in the main pulse, which is responsible for completely stripping the electrons from carbon.

5.6 Summary

In this chapter, we presented the CVI lines-dominated spectra of ethane plasmas created by a 100 fs, 200 mJ pulse, focused by an off-axis parabolic mirror into a focal spot of 3.6 μm diameter and peak intensity reaching $10^{19}$ W/cm$^2$. Similar spectra, with weaker dominance of CVI lines, were acquired using 25 ps pulses despite the different
ionization mechanisms. The role of initial gas density in pressure broadening was identified. Then we improved the contrast of the main pulse by inserting two RG850 saturable absorbers before the final amplifiers of the Thales Alpha system. The suppression of the pre-pulse not only enhanced the dominance of CVI lines in the observed spectra, but also increased the relative populations of the upper levels in n-1 transition in CVI ions. By manipulating the initial gas density, we also observed a clear indication of a higher population of level n=5 than that of n=4 in CVI [5.20]. The acquired spectra resembled the earlier LiIII spectra which led to the generation of high gain and lasing to ground states when a longer plasma channel was produced. Preliminary attempts to extend the plasma with promising conditions for lasing to ground state in CVI also yielded encouraging results, demonstrating a well-confined propagation of 25 ps pulses inside the plasma waveguide produced by a 200 ps, 190 mJ Bessel beam. An upgrade of the Positive Light laser system, aiming to deliver more energy for plasma waveguide formation, is in progress.
Reference:
Chapter 6

Conclusions

6.1 Summary

The major results and progresses in this dissertation can be summarized as follows.

We first investigated the propagation of sub-picosecond laser pulses, both experimentally and numerically, in optically ionized hydrogen and nitrogen gases at an intensity of $4 \times 10^{16}$ W/cm$^2$. Strong ionization-induced refraction resulted in much shorter propagation distance in nitrogen than in hydrogen. Moreover, forward Raman scattering and ionization scattering were also identified, in numerical modeling, as causes of the disruption of the pulse propagation.

Then we described the development of the modified igniter-heater technique to produce two types of high-quality plasma waveguides. The combination of a 200 ps Bessel beam and a 10 ns spherical beam successfully created plasma waveguides of the hollow-pipe structure with over 450 $\mu$m diameter and $10^{19}$ cm$^{-3}$ on-axis density. Plasma waveguides of less than 10 $\mu$m diameter were demonstrated for application in XRL research using the combination of a 100 fs pulse and a 200 ps pulse, both of which were Bessel-like. The axial corrugation of plasma waveguides were effectively suppressed or even eliminated experimentally when the 100 fs igniter was introduced or hydrogen was mixed with the ethane or nitrogen gas jet. These plasmas will play a critical role not only in soft x-ray lasers, but in other applications as well, such as the Raman amplification.
The spectroscopic features of ethane plasmas resulted from the interaction between 100 fs or 25 ps pulses with the ethane gas jet was studied in depth. With the suppression of the pre-pulse and its associated pre-plasmas using the RG850 saturable absorbers, we produced CVI-dominant plasmas with higher emission intensity from the 3-1 transition than that from 2-1 transition, indicating possible 3-2 population inversion in CVI ions. Particularly for 100 fs pulse, we were able to change the relative populations of the upper levels (n=3, 4, 5) of the emission lines by adjusting pumping pulse energies and the plasma densities. The astonishing resemblance to the LiIII spectra, which were later found to produce high gain in lasing to ground states, lent us confidence for obtaining lasing to ground states of CVI and CV ions.

Preliminary experiments were also conducted to study the propagation of powerful pulses in plasma waveguides through shadowgrams. Elongated plasmas with boosted density were found when the 25 ps pulse was focused at the entrance of the pre-formed plasma waveguide. The radiation was well confined within the waveguides all the way through the gas jet, instead of a disruption long before arriving at the center of the jet if no waveguide was pre-formed.

6.2 Future direction

Based on the achievements and problems during the previous efforts, the direction for future work can be divided into technical and conceptual categories.

On the technical side, the most helpful work is the improvement of Positive Light that delivers pulses for pre-formed plasma waveguides. During the preliminary experiments on pulse propagation, the formation of plasma waveguides was so irreproducible that no
consistent spectra could be taken. Similar improvement can also be made on the Thales Alpha system that delivered our main pulse for soft x-ray lasers, so we can have more flexibility in choosing proper parameter windows. Other than improving the output of the laser, we can also reduce the energy losses along the beam path. The 20%-30% loss on the last reflection mirror of the main pulse could be further reduced if we replace the mirror by a thin film polarizer provided that the polarization of the pulses is compatible.

Another helpful improvement of the setup is convenient monitoring of the 100 fs pulse profile at the focus. Currently we need to open the vacuum chamber to set up the imaging system. It will be of great help if a mechanical arm, controlled electrically outside the chamber, can be installed. Therefore, we can check the beam profile in vacuum without interrupting the experiments.

Further reduction of scattering light on the CCD will be extremely beneficial in detailed study of the spectra. The noise at the short wavelength region in current setup is sometimes quite overwhelming, in which case our ability to study spectral lines in that region is severely impaired. Additional pinholes of only a few µm will be particularly helpful because the 800 nm radiation will be greatly diffracted while the soft x-ray signal of much shorter wavelength can freely pass through.

Motorized translation of the axicon assembly will improve the efficiency and precision of our alignment. This improvement is highlighted in the injection of powerful pulses into the plasma waveguides since motorized translation will enable us combine the two ionizing pulses without opening the vacuum. A relevant improvement is a longitudinal shadowgram system which directly monitors the spatial overlapping of the two pulses. The green beam after pumping the last amplifiers of Thales Alpha system can be brought
out as the probe beam from the dumper. Its advantage lies in the fact that this green beam is synchronized well with the main pulse.

In the conceptual category, on top of our priority list is the development of an elaborate numerical model that simulates the propagation of ultrashort pulses in partially-ionized plasma waveguides. We would like the model to start from the ionization of Bessel beams, and compare the results with experiments. Then we can deal with the propagation of powerful pulses in plasmas whose initial condition is verified in both experiments and the previous step of simulation, taking into account the additional ionization, and as many nonlinear processes as possible. It is beyond any doubt to provide valuable direction for future experiments. Currently, we are in collaboration with Dr. Phillip Sprangle and Dr. Dan Gordon in Naval Research Laboratory on this project.

Adding hydrogen is another important attempt to create more favorable plasma conditions for population inversion and gain. Since the intermediate processes between adding hydrogen and gain creation are complex and entangled, it will be very helpful to set up a diagnostic system for plasma temperatures. Experimental establishment of a direction connection between adding hydrogen and the reduction of plasma temperature will help us in designing subsequent experiments. Equipped with all these improvements, within a quite compact system, we expect to see a bright future about soft x-ray lasers operating in the “water window”.