Tabletop Ultrabright Kiloelectronvolt X-Ray Sources
from Xe and Kr Hollow Atom States

BY

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THESIS
Submitted as partial fulfillment of the requirements
for the degree of Doctor of Philosophy in Physics
in the Graduate College of the
University of Illinois at Chicago, 2013.

Chicago, Illinois

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This thesis is dedicated to my elder brother Mr. Poopalasingam Gopi and to the memory of my younger brother Poopalasingam Poneeshan.
ACKNOWLEDGEMENTS

First and foremost, I offer my sincerest gratitude to my supervisor, Professor Charles Kirkham Rhodes, who provided me a broad platform to learn diverse experimental and administrative skills during the ultra-beam project. His brainstorming ideas and guidance resolved most of the experimental problems and led to the success of this thesis. I also thank the members of my graduate committee Professors Charles Kirkham Rhodes, W. Andreas Schroeder, Wai-Yee Keung, Alex Borisov, and Robert J. Gordon, for their guidance, suggestions, and for their golden time. I thank Physics department head Professor David Hofman for supporting me in numerous ways through out my PhD program in UIC.

I could not find words to express my sincere thanks to Dr. James Longworth for valuable discussions, guidance, and literature searches. This thesis would not have been possible without his reviews, corrections, and recommendations. Thanks Jim for all the helps. I am so lucky to have you in the lab.

I would like to thank my parents Mr. Ponniah Poopalasingam, and Mrs. Manomany Poopalasingam for their love, support, and care. I am extremely blessed to be your son and I will be remembering forever all of your sacrifices and hard works to educate all of your children. Further, I would like to thank my brothers Gopi, Moorthy, and Sivakumar, my sisters Gomathy, Pooma, and Selvam, my nephews Kiruthikan, Mathuran, Soruban, Vishnu, Piradeep, Kabilan, and Sandeep, nieces Mathusha, Rahavi, Kaja, Kayali, Mayuri, Paraneka, Jasotha, and Abirami for their love and support to walk through this journey. Many thanks go to my mother-in-law Mrs. Karunadavei Velmurugu, sister-in-laws Ms.
ACKNOWLEDGEMENTS (continued)

Jegathambigai and Ms. Vimalambigai for all of their prayers to my successes.

Special thanks go to my lovely wife Yoga and daughter Biranavi Ambigai for their patient, numerous supports, and encouragements to pursue this degree. Without my wife Yoga’s encouragement, supports, and hard works I would not have finished the degree. My wife and daughter provided me such a comfortable environment to focus on designing and spending enough time for machining mechanical parts to make the laboratory instruments ready for challenging experiments. Most of parts of this thesis were written while I take care of newborn baby Biranavi Ambigai. My baby girl gave me such an extremely comfortable environment to complete this thesis.

My sincere thanks goes to Mr. Sivaparan, Ms. Raji and Sivajan for providing me such a hospitality to familiarized with US culture and environments immediately after my arrival to USA. Thanks Raji for your fresh cooked food. Thanks Sivaparan for your supports in numerous ways. Sivajan your friendship made me to pass though my early homesick period.

I thank my fellow lab mate Mr. John McCorkindale for the sleepless nights we were working together before deadlines, and for all the fun we have had in the last two years. I thank my former lab mates and friends Dr. Ervin Rácz, who helped me to design and built XRIM laboratory and Dr. Shahab Firasat Khan for developing LabVIEW code to recorded single-shots data. It was fun to work with you guys.

I would like to thank physics department machine shop supervisor Mr. Kevin Lynch for always giving me priorities to my projects. I also like to thank Mr. Richard Frueh, Mr. Richard Dojutrek, Mr. Robert Kurdydyk, Kurt, and Tony for providing me your
ACKNOWLEDGEMENTS (continued)

instrumental skills to build the laboratory. I like to thank Mr. Derrick Stanley for placing purchasing orders always in front at short notice without any hesitant. I really appreciate Mr. Randy Carlson and Mr. John Quicksilver for providing technical assistant to operate Prometheus amplifier, and Mr. David Virgillito for providing technical assistant to operate Hurricane laser. Last but most I like to thank Melissa Mattingly, Melodie Shaw, and James Nell for treating me as a friend and helping me in various ways from 2003 to 2013.

P. Sankar
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<tr>
<td>AMP</td>
<td>Amplifier</td>
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<tr>
<td>ASE</td>
<td>Amplified Spontaneous Emission</td>
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<tr>
<td>BET</td>
<td>Beam Expanding Telescope</td>
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<tr>
<td>BM</td>
<td>Big dielectric beam steering Mirror</td>
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<tr>
<td>BNL</td>
<td>Brookhaven National Laboratory</td>
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<tr>
<td>CCD</td>
<td>Charge Coupled Device</td>
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<tr>
<td>CPA</td>
<td>Chirped Pulse Amplification</td>
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<td>DA</td>
<td>Direct Amplification</td>
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<td>DGSN</td>
<td>Double Gas Sonic Nozzle</td>
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<tr>
<td>DNA</td>
<td>DeoxyriboNucleic Acid</td>
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<td>DOC</td>
<td>Dynamical Orbital Collapsed</td>
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<td>FSMOS</td>
<td>Focal Spot Monitoring Optical Setup</td>
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<td>HHG</td>
<td>High-order Harmonic Generation</td>
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<td>ICS</td>
<td>Inverse Compton Scattering</td>
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<td>IR</td>
<td>Infra Red</td>
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<tr>
<td>ISO</td>
<td>International Organization for Standardization</td>
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<td>LASER</td>
<td>Light Amplification by Stimulated Emission of Radiation</td>
</tr>
<tr>
<td>LCLS</td>
<td>Linac Coherent Light Source</td>
</tr>
<tr>
<td>LLG</td>
<td>Laser Laboratorium Göttingen</td>
</tr>
<tr>
<td>LLNL</td>
<td>Lawrence Livermore National Laboratory</td>
</tr>
<tr>
<td>LU</td>
<td>Lumenera Corporation</td>
</tr>
<tr>
<td>MASER</td>
<td>Microwave Amplification by Stimulated Emission of Radiation</td>
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<tr>
<td>NA</td>
<td>Numerical Aperture</td>
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<tr>
<td>OAP</td>
<td>Off Axis Parabolic mirror</td>
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<tr>
<td>OTBI</td>
<td>Over The Barrier Ionization</td>
</tr>
<tr>
<td>PI</td>
<td>Princeton Instruments</td>
</tr>
<tr>
<td>PPPL</td>
<td>Plasma Physics Laboratory, Princeton University</td>
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<td>QPM</td>
<td>Quasi Phase Matching</td>
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LIST OF ABBREVIATIONS (continued)

RA   Regenerative Amplifier
RMS  Root Mean Square
SHG  Second Harmonic Generation
SLAC National Accelerator Laboratory (Stanford)
TG FROG Transient Gating Frequency Resolved Optical Gating
THG  Third Harmonic Generation
TS   Thomson Scattering
UIC  University of Illinois at Chicago
UV   Ultra Violet
XFEL X-ray Free Electron Laser
XRIM X-Ray Micro-Imaging and bioinformatics laboratory
SUMMARY

Wilhelm Conrad Röntgen discovered x-ray in 1898. Today x-rays play crucial roles in medical field as well as in industrial field. However, currently available tabletop traditional x-ray sources such as the one in the dental-field have low quality skillful brightness, which limits its resolution and accuracy.

Accelerator-based ASE x-ray sources such as x-ray free-electrons laser and synchrotron operate in underground large-facilities currently generates partial coherent and moderate pulse-length ultrabright x-rays. The x-rays from these larger facilities provide direct visions of electronic and structural changes of biology, atomic, and nanoscience at molecular level; scientists have never had such a window through which to explore the nanoworld. However, number of applications of such underground larger facilities such as medical and industrials, has been hampered by their size, complexity, and cost. This has set a goal of demonstrating x-ray source with enough brightness for potential applications in an often-called tabletop compact x-ray source that could be operated in university laboratory or hospitals.

The research goal of the research presented in this thesis is to realize a compact-size ultrabright, tabletop x-ray source device with unique characteristics that make it complementary to x-ray free-electron lasers and synchrotron x-ray sources. The compact-size of such x-ray sources will make them ideal for installation in university, hospitals, and industrial laboratory settings and will be accessible to ordinary peoples.

Realizing coherent ultrabright x-ray source in the x-ray wavelength at laboratory-scale is facing extremely challenging practical problems, as it required higher pump-
power, scarcity of available target-medium, and optics in x-ray wavelength regime. XRIM is a pioneer in the field of generation of ultrabright x-ray source and applying unique innovative ideas to resolve the challenges of realizing laboratory-scale ultrabright kiloelectron Volt (keV) x-ray sources. In order to acquire required pump-power for lasing action at laboratory-scale, a brand-new ultra-fast hybrid tabletop KrF* pump-laser, laboratory, and experimental chamber is designed and built to deliver 1 watts/atoms level pump-power to target-medium at 0.1 Hz repetition rate. Accomplished 2 µm spatial pointing stability of pump-laser on target-medium confirms the success of design of pump-laser system and laboratory. A hybrid Double Gas Sonic-Nozzle (DGSN) is custom designed to produce optimal target gas clusters inside vacuum chamber. Interaction of pump-laser with target-clusters produce inversely populated hollow-atom states, which later coherently collapse to the inner-shell and generate x-ray radiation in the keV wavelength regime. Self-focused KrF* pump-laser beam forms a dielectric waveguide and self-channel the generated x-ray radiation in the direction of propagation of the pump-laser beam. All of calibrated diagnostics are redesigned to upgrade their performances and reliability to record single-shot data during the interaction of pump-laser with target-medium. Typically, each recorded data file includes (1) the 248 nm pump-laser energy (mJ), (2) the target gas sonic nozzle pressure (psig), (3) target gas temperature (K), (4) triple x-ray source images captured by triplet-aperture x-ray camera, (5) a transversely observed spectrum, (6) an axially observed spectrum, and (7) transversely captured Thomson scattering image.
SUMMARY (continued)

A decade of design, construction, research work, and tireless handwork realized a Xe x-ray source in Xe M-shell wavelength range (1.2-1.6 nm) and a Kr x-ray source in L-shell wavelength range (600-800 pm). The system is mounted upon 3 optical tables (5’x12’) along with two KrF amplifiers operates at 0.1 Hz repetition rate. The design and engineering is capable of delivering ultrabright keV x-ray beam within two hours time need for warming-up pump-laser, amplifiers, and diagnostics. Experimentally estimated lower bound brightness value for the realized Xe and Kr x-ray sources is $10^{27}$ photons s$^{-1}$ mm$^{-2}$ mrad$^{-2}$. This brightness value is greater than average brightness ($10^{22}$ photons s$^{-1}$ mm$^{-2}$ mrad$^{-2}$) of available x-ray free electron lasers and synchrotron x-ray sources [14], [15], and [16].

Innovative diagnostics design, increased instrumental sensitivity and dynamic range approximately 100-fold greater compare to previous diagnostics and led to new discoveries. More than 3 terabytes data and over two hundred thousands single-shot data files are recorded from the experiment performed in the newly build system. Although much has been learned, this thesis is presenting only few key findings, which are summarized below.

Aperture x-ray camera experimental findings show that x-ray source morphological types are varies shot by shot. Modus operandi of the system has complete control of selecting desired morphological type. Recorded Xe M-shell and Xe L-shell radiation images simultaneously by the triplet-aperture x-ray camera further shows that the Xe M-shell radiation is coupled to the Xe L-shell radiation and predicts anomalous photons ma-
SUMMARY (continued)

ter coupling taking place during the experiments. This anomalous coupling favors the mission of generation of shortest ultrabright x-ray source.

Thomson scattering experimental data show that generated x-ray radiations are trapped inside a dielectric waveguide and self-channeled in the direction of propagation of pump-laser. Combination of theoretical and experimental estimations predict that diameter of the waveguide is in the order of x-ray source center wavelength. Further, the Thomson scattering data reveal that length of the self-channel is 30 times longer than Rayleigh range of pump-laser focused spot on target clusters. X-ray energy flow estimations in the transverse and axial-direction validate the x-ray self-channel formation.

Comparison of single-shot transverse-spectra collected from the beginning of self-channel to transverse spectra collected from the end of self-channel shows enhancement of x-ray signal strength at the end of self-channel. The Kr L-shell spectroscopic analysis shows that Kr L-shell x-ray source has two dominant x-ray emissions lines, one is enhanced Kr 3d $\rightarrow$ 2p transition at 0.67 nm and the other is the enhanced Kr 3s $\rightarrow$ 2p transition at 0.75 nm. The average lower-bound energy of these two x-ray emission lines at a distance of 3.5 cm from the center of the x-ray sources are in the order of 8 µJ energy with attosecond pulse-width. Simultaneous comparison of single-shot axial and transverse-spectra at various on-axial positions shows that in the direction of propagation of pump-laser, the transverse-spectra become faint while axial-spectra develop stronger.

DGSN is designed to trap the target gas into a smaller volume with definite sharp boundaries by spraying secondary low-Z gas at target gas annular region. It is expected that the action of DGSN operation to optimize cluster density and to reduce x-ray attenu-
SUMMARY (continued)

ations during the propagation of x-rays through unionized target gas column. Comparison of axial x-ray energy estimations with and without operation of secondary gases shows that the operations of secondary gas double the average x-ray energy and supports self-channel formation. It is further shows that operation of DGSN could produce 100 µJ Xe M-shell x-rays at a distance of 3.5 cm from center of the x-ray source.

Realized x-ray sources shows evidences for laser-like natures such as coherent and directionality. Further research works are necessary to confirm these observations. If the sources are succeed in these tests, the realized ultrabright keV x-ray source will be a perfect candidate for lithographical, high-precision holographic micro fabrication, non-destructive testing with phase-sensitive projection imaging applications and will be a laboratory-scale brightest keV x-ray source to date.
1. INTRODUCTION AND MOTIVATION

1.1 A Brief Thesis Introduction

Albert Einstein, the father of relativity, once said, "Look deep into nature, and then you will understand everything better". Today available higher resolution tool to look deep into matters and living thing is an x-ray source. Although the available tabletop x-rays sources of the 20th century, such as the ones used for medical or dental x-rays are tremendously useful for medical diagnostics and industry, a major disadvantage is that they produce poor brightness; a factor determines the resolution of the source. X-Ray Micro Imaging and Bioinformatics Laboratory (XRIM) team lead by Albert A. Michelson Professor Charles Kirkham Rhodes at department of physics, University of Illinois at Chicago (UIC) is pioneer in the research field of developing a tabletop x-ray source with possible higher brightness. This thesis is presenting research work performed at XRIM for developing such a revolutionary tabletop ultrabright directional x-ray sources that can produce x-rays in 0.5 - 1.6 nm wavelength range.

First chapter of thesis, introduction and motivations, goes back to 18th century and begins with discovery of x-rays and then it discusses limitation of the traditional x-rays for their applications. Afterwards it discusses the revolutionary applications of coherent x-ray sources as well as difficulties in generation of coherent x-ray sources. After explained the difficulties in generation of coherent x-rays, it discusses available methods for generation of coherent x-ray source. Finally, this chapter briefly elaborates how XRIM approaches the challenges to realize tabletop ultrabright directional x-ray sources. It is the motivation of the research presented in this thesis.
The second chapter, experimental method, and apparatus, begins with presenting experimental method to generate ultrabright x-ray sources. The section apparatus presents apparatus design, experimental setup, and alignment procedures for successful generation, optimization, and characterizations of ultrabright x-ray sources at XRM.

In third chapter, theoretical framework, already existing theories are borrowed to estimate experimental parameters and to explain experimental observations. High-intensity focal-spot waist-size and target-medium parameters such as target cluster-size at focal-plane are estimated in first two sections. After ward, various ionization mechanisms take place during the experiments are discussed to understand the physics behind the experimental observations recorded by diagnostics.

In the fourth chapter, results and discussions, single-shots experimental data collected by calibrated diagnostics are systematically discussed under each diagnostics. First section of chapter 4 is focused to verify the realized x-ray sources are in the kiloelectron Volt (keV) x-ray wavelength regime. Remaining sections of chapter 4 are investigating realized Xe, and Kr x-ray sources to estimate single-shot x-ray source energy, brightness, x-ray self-channel length, and radius. A hybrid Double Gas Sonic Nozzle (DGSN) is custom designed to spray primary single target-gas at center and low-Z secondary gas in the primary target-gas annular area to optimize target-cluster density while reducing x-ray attenuation during its propagation through unionized target-gas column. End of chapter 4.4 presents experiments performed to verify design objectives of DGSN.

Final and last chapter five summarizes realized x-ray sources characteristic parameters and conclude the research work presented in this thesis.
1.2 Discovery of X-rays

In the historical evolution of exact sciences, the nineteenth century is a crucial period. Brilliant discovery of polarization of light by Malus, study of magnetic-field by Faraday, formulation of mathematical expression for electromagnetic-field by Maxwell, formation of basic-laws of thermodynamics by Clausius and Maxwell, theories of gases by Maxwell and Boltzmann, observation, explanation of cathode rays, and discovery of electrons by J. J Thomson, are some of the revolutionary theoretical and experimental discoveries of nineteenth century. These discoveries formulated fundamental concepts for the exact-sciences [1]. Among these discoveries, mysteries of the cathode rays remained unsolved until the last quarter of the 19th century. During the last quarter of the 19th century, many experiments were performed to understand the cathode ray. The experiments not only solved mysteries of cathodes rays but also resulted to groundbreaking discoveries such as x-rays (1895), electrons (1897), and explanation of electrical current. The brief history of the great discovery of the x-rays, which is the center of attraction of this thesis, is presented below.

In the October of 1895, professor of physics and the director of the physical institute of the University of Wurburg Wilhelm Conrad Röntgen (1845-1923) had assembled equipment for the taking up work on the hotly-contest of 19th century in the subject of cathode rays. He fully enclosed the tube in a light-tight cardboard box to make sure to observe a very faint luminescence. He placed barium-Platino cyanide screen on a table at a considerable distance from the tube. He observed flash of fluorescence every time a discharge of the induction coil went through the tube. The flash could not be due to cathode rays because these cathode rays would have been fully absorbed either by the
glass-wall of the tube, or by the air. Röntgen, in a breathless period of work between November 8 and the end of the year 1895, convinced himself of the reality of his observation, which at first he found hard to believe. He soon concluded that some thing, the unknown x, which traveled in a straight-path from the spot where the cathode ray in the tube hit the glass wall, caused the fluorescence. Röntgen later named the unknown rays as x-rays [1].

Röntgen placed various objects between the tube and screen and found that the screen still fluoresced but with different intensities depending on the material being used. Then, in a heart-stopping moment, he chanced to pass his hand through the beam. As he looked at the screen, the flesh of the hand seemingly melted away, projecting only the outlines of the bones. The hand was intact, unharmed but on the screen, only the bones showed up. That observation later gave birth to science of medical radiology. A few days later, Röntgen made a photographic image of his wife’s (Frau Röntgen) hand, using the new rays instead of light for the exposure. Again, only the bones showed, this time on a permanent record, which others could see and believe. Röntgen published his accidental discovery in the proceedings of the physical medical society of Würzburg on December 28, 1895 with title "On a New Kind of Rays". In 1896, he accepted the Rumford gold medal of the royal society and in 1901; he would be the first to receive the Nobel Prize for physics [1]. X-rays found its first application in the field of medicine just few months after the discovery and now the great discovery plays crucial roles in various fields.

The terms “x-rays”, today, can mean a source of electromagnetic radiation between 0.01-10 nm of an electromagnetic spectrum, which is presented below in figure 1.1.
The lower and upper-boundaries of x-rays regime in the electromagnetic spectrum are not well defined. The longest-wavelength of the x-ray is overlapped with the shortest-wavelength-edge of the Ultra-Violet (UV) light and shortest-wavelength of x-ray is overlapped with longest-wavelength of gamma rays. According to its wavelength x-rays find various applications. The following section 1.3 briefly discusses the applications of traditional x-ray sources.

1.3 Applications of Traditional X-rays

Röntgen by presenting an x-ray photograph of his wife’s hand to the Würzburg Physical and Medical Society in January of 1896 laid first seed to the practice of radiology. A month later, a German doctor used x-ray to diagnose sarcoma of the tibia in the right leg of a young boy [2]. It was the very first application of x-rays in the field of **

** Courtesy of the Advanced Light Source, Berkeley Lab.
medicine. Nowadays, the uses of x-rays are found in a variety of applications in the field of medicine as well as in all other fields. Some example medical applications include

- X-ray radiography, a technique to find orthopedic damage, tumors, pneumonias, and foreign objects.
- Mammography, which uses x-ray to early detection of breast cancer.
- Computer Tomography (CT) to produce cross-sectional images of human body.
- Fluoroscopy, a tool to dynamically visualize the body and determine where to remove plaque from coronary arteries or where to place stents to keep those arteries open.
- Röntgen stereophotogrammetry, which uses x-rays to track movement of bones based on the implantation of markers.
- Radiation therapy for treatment of cancer.

Some the applications of x-rays in the fields other than field of medicine are presented below.

- Industrial radiography uses x-rays for inspection of industrial parts, particularly in welds.
- X-ray crystallography, a technique reveals the nature of lattice of crystals.
- Fiber diffraction, a procedure used by Rosalind Franklin to discover the double-helical structure of DNA [3].
- X-ray photoelectron spectroscopy is a chemical analysis technique relying on the photoelectric effect, usually employed in surface science.
- X-ray lithography, is a process used in electronic industry to selectively remove parts of a thin film for the semiconductor industry.
Security luggage scanners use x-rays for inspecting the interior of luggage for security threats.

Although the traditional x-rays find numerous applications, its accuracies are often limited by characteristics of traditional x-rays employed. Traditional x-rays lack of two particular characteristics that would be ideal for applications. The desirable characteristics are coherent nature and pulse-width of the x-ray source.

Coherent nature includes temporal-coherence and spatial-coherence. The temporal-coherence describes the correlation or predictable relationship between signals observed at different moments in time. In other words, the temporal-coherence describes how monochromatic the source is and it characterizes how well a wave can interfere with itself at a different time. Spatial-coherence describes the correlation between signals at different points in space.

The pulse-width could be defined as time interval between the leading edge and trailing edge of a pulse at a point where the amplitude is 50 percent of the peak value. A mathematical expression for the pulse-width is presented in the chapter 4. An x-ray source with possible shortest pulse-width will be a suitable candidate for viewing fast moving objects and for some of medical applications such as cancer radiation therapy. The diagram 1.1, below, describes the necessary pulse-width needed for resolving different physical phenomena.
Currently available tabletop traditional x-ray sources such as the one in the dental-field, in the medical, and in the industrial-field have very poor coherent nature and long pulse-length. Realization of tabletop x-ray source that produces coherent and ultra-short pulsed x-ray will make revolutionary implication in science. New advances in atomic and optical physics are able to create brilliant bursts of x-ray beams with laser-like properties. These bright, directed x-ray beams can be focused to the size of a virus and are fast and bright enough to capture the complex dance of atoms within molecules or even faster than fleeting motion of electrons within atoms and molecules. These extreme strobe lights, with x-ray vision, will provide a direct-view of the electronic and structural changes that govern biology and nanoscience at the molecular level; scientists have never
had such a window through which to explore the nanoworld in ordinary laboratories [4]. The following section 1.4 elaborates such coherent x-ray source.

1.4 Coherent X-ray Source

An enlightening application of x-ray in medical field is imaging of living tissues and cells. In particular, by using x-rays one should be able to study macromolecules in living tissues, thereby gaining crucial insights into cellular functions. The reason this has not yet been done is that the index of refraction of low atomic number (Z) elements is close to unity through out the x-ray region, and therefore targets containing low-atomic number elements offer poor image contrast when illuminated with ordinary traditional x-ray sources. A source of coherent x-ray with short enough pulse-length, on the other hand, would make phase contrast microscopy possible, which would yield contrast sufficiently high to allow the study of biological macromolecules. Further, a coherent source would, in fact, permit one to make three-dimensional hologram of important biological structures such as the DNA in a cell nucleus. Additionally, by using a short x-ray pulse (about 10\(^{-15}\) sec), in material science, one could “freeze” molecular vibrations, which will give possibility to watch, in slow motion, thermal vibrations-even shock wave compression in various materials [5].

It is clear from all the facts above that a coherent x-rays would have revolutionary implications for many areas of biology, chemistry, physics, and material science. What then, are the prospects for generating these coherent x-rays? An answer to this question is briefly presented in following section 1.5 after the discussion of brief history of coherent sources presented below.
History of manmade tabletop coherent sources begins with the discovery of Microwave Amplification by Stimulated Emission of Radiation (MASER) invented by Columbia University Professor Charles H. Townes with the help of Herbet Zeiger and James Gordon in the early 1950. Since wavelength of microwave maser is in the order of centimeters (cm) (10,000 longer than visible-light), by middle of 1950 maser research started to loose interest and researchers were started to focus on different ideas for generation of shorter-wavelength coherent sources.

Theodore Maiman succeeded on May 16 1960 at Hughes research laboratory in making first working coherent and monochromatic light-source with the wavelength 694.3 nm, which is $10^{-7}$ times smaller than wavelength of maser. Maiman used his knowledge of ruby masers and adopted Gordon Gould, a Columbia graduate student, idea about optical pumping for his dissertation. The device is known as Light Amplification by Stimulated Emission of Radiation (LASER).

The Maiman’s traditional quantum lasers consists of three basic components; lasing-medium with at least three energy levels, an optical-resonator, and pump-energy for successful lasing action. The basic concept of the traditional quantum laser is presented below in the diagram 1.2.
The successful demonstration of working ruby laser encouraged laser scientist to pursue research work to realize lasing action at shorter and shorter wavelengths and initiated “race for short wavelength lasers”. The “race for shorter wavelength lasers” accelerated up to UV wavelength regime and then slowed down rapidly at x-ray wavelength regime and faced torturous difficulties and challenges as presented below in following section 1.5.

1.5 Challenges of Realizing X-ray Lasers

Discussion of challenges of realizing X-ray lasers begins with the definition of X-ray lasers.

An X-ray laser defined as a source, which delivers quasi-monochromatic, coherent, or partially coherent photons in the spectral range shorter than a few tens of nm [6].

There have been so many different ideas proposed for realizing x-rays lasing. Initially, it was though that X-ray laser would be a direct implementation of traditional
quantum lasers. However, implementing the traditional laser ideas for realizing X-ray laser faces serious practical challenges. These excruciating challenges are described below followed by clever solutions sought by XRIM to resolve the challenges.

1.5.1 Challenge 1: Lasing Medium

The central part of traditional laser is lasing-medium. However, matter is highly opaque at all x-ray wavelengths due to photoelectric absorption, which is limiting the availability of lasing-medium at x-ray regime. It was suggested that plasmas would be one of the suitable candidate for the lasing-medium at x-ray wavelengths. In the XRIM, Xenon (Xe) and Krypton (Kr) atomic-clusters are the main medium for the generation of ultrabright x-ray sources. The complete details of these lasing-mediums are presented in chapter 3.

1.5.2 Challenge 2: Cavity Mirrors

One of the primary obstacles in developing a laser that emits electromagnetic radiation at x-ray wavelengths is that the available short-wavelength mirrors have very low reflectance. This makes it difficult to achieve gain in an oscillator cavity composed by mirrors at its ends. Therefore, the lasing at x-ray wavelengths must probably be operated without external mirrors, due to the low reflectivity of materials at the x-ray wavelength regime.

There have been so many different ideas proposed to overcome the challenges to generate and sustain population inversion for lasing action in the x-ray spectral region. In 1967, two Bell telephone laboratories, Inc scientists M.A. Duguay and P.M. Rentzepis
proposed a completely different idea for sustaining population inversion without any external-mirrors for realizing coherent source in x-ray wavelength regime. The idea is; ejection of electron preferentially from inner-shell UV and x-ray wavelengths [7]. It is possible that photons with proper energy range impinging upon an atom could preferentially remove inner-shell electrons and thus induce population inversions with respect to outer-shell electrons. Such a scheme leads to transient lasing at very short wavelengths [7]. As an example when an electron has been ejected from, say, the K-shell of an atom, the residual ion is left in an excited state that can later decay by virtue of an L-shell electron, spontaneously, dropping down into the K-shell. This decay process will emit photons in x-ray wavelength regime. The residual ion with inner-shell vacancies is termed, in this thesis, as “hollow-atom”.

X-ray radiation from a single “hollow-atom” is not sufficient to produce an intense x-ray radiation. However, simultaneous emission of x-rays from collection of “hollow-atoms” would be able to produce intense coherent x-ray radiation. Even before the invention of the first working lasers, 1953, Robert H. Dicke, physics professor at Princeton University detailed in his theoretical work by treating radiating-gas as a single quantum mechanical-system showed that the spontaneous emission of radiation in a transition between two-levels leads to the emission of coherent radiation [8]. Therefore, one of potentially very effective methods of producing an extremely bright source of x-ray radiation would be to generate a controlled population inversion in the inner-shells of high-Z atoms. In other words, producing specific species of “hollow-atoms” would be one of potential solution for unavailability of cavity mirrors at the x-ray wavelength
regimes. The primary pump-mechanism in XRIM for producing ultrabright x-rays is “hollow-atoms” mechanism.

1.5.3 Challenge 3: Pump Power

Another barrier for lasing at x-ray wavelength is that very high-pump-powers are required to create and maintain the necessary population inversions for the lasing action. This is due to very short excited-state lifetimes and the higher-transitions energies involved [5]. A crude analysis shows that that the pump-power required for a given amplification must be roughly proportional to the inverse fourth power of the wavelength ($\lambda^{-4}$) [10]. Further, a simple relation between gain $G$, wavelength $\lambda$, and population inversion $\Delta N_{inv}$ ($\sim$ pumping-power) is described in the reference 10 and presented below by equation 1.1.

$$G \sim \lambda^4 \Delta N_{inv}$$  \hspace{1cm} (1.1)

According to the equation 1.1, in order to decrease the lasing wavelength from 10 to 1 nm at constant gain $G$, the pump-power must be increased approximately by factor of $10^4$. The following simple example illustrates the scenario clearly. In order to maintain a vacancy (with life time of one femtosecond) in the inner-shell of one copper atom, an 8 k eV photon needs to be absorbed $10^{15}$ times per second; it takes about 1 watts per atom to maintain this K-shell vacancy. To put this power requirement into perspective, the entire United States energy consumption of the year 1994 would suffice only to keep only 0.1 nano-gram of copper pumped up continuously [9]. Therefore, concentration of power (it is popularly know as “1 watts per atom”) can be seen as the central problem in X-ray
laser design. Following paragraph describes how a powerful traditional pump-laser produces enough power (1 watt per atom) for lasing in x-ray wavelengths.

A building-size Nova fusion lasers at Lawrence Livermore National Laboratory (LLNL) is the one first reached the required pump-power (in beginning of 1980) for lasing at x-ray wavelength. The Nova was able to fire only about six-times a day because of very short-pulses overheated the glass amplifiers and time needed to cool down glass amplifiers. The laser chirped pulse amplification (CPA), developed in the late 1980s, gets around that problem by expanding a very short-pulse before it travels through the amplifiers and then compressing it to its original duration before the laser beam is focused on a target. CPA was originally introduced as a technique to increase the power in radar in 1960. In the mid 1980 Gérard Mourou and Donna Strickland at the University of Rochester invented CPA for lasers. Since the CPA is combined with lower-energies, the pulses do not overheat the glass amplifiers, so the system at LLNL was able to fire several times a day. Nowadays the CPA technique made it possible laboratory-size terawatt ($10^{12}$) and petawatt ($10^{15}$) pump-lasers for achieving enough pump-powers for tabletop X-ray lasers.

Even though the CPA technique made is possible to reach required pump-powers for lasing at x-ray regime, maintaining the population inversion at laboratory-scale is the torturous challenge for the development of tabletop X-ray lasers. A successful solution attempted at XRIM by seeking new modes of energy delivery and power concentration that established a more favorable scaling relationship between the magnitude of gain produced and excitation power required for x-ray amplification. There are two physical phenomena are assumed to play a role in producing large-power compression in the
experiments; a novel mode of multi-photon excitation of clusters and a relativistic regime of high intensity pulse propagation in plasma. A brief description is provided in chapter 3.

According to the wavelength range of x-ray, excitation mechanism, the basic principles of generations of coherent x-ray sources, and applications varies. In the following section 1.5, the X-ray lasers according to their operating wavelengths, are divided into four-group, soft, water-window, kilovolts and hard x-rays.

1.6 Classifications of X-ray Lasers

The X-ray lasers, according to the wavelengths regimes divided below into four-categories and presented below.

1.6.1 Soft X-ray Laser

First regime is known as soft X-ray laser. A compact high repetition rate tabletop soft X-ray laser is a promising candidate for manufacturing integrated circuits in large-scale using projection lithographical techniques. Soft X-rays lasers are not suitable for medical imaging but they are ideal tool for probing and imaging the expansion of high-energy density plasmas. In 1985, Matthew et al at LLNL published first demonstration of a soft x-ray lasing at wavelengths in the range of 20.63 and 20.96 nm from the Neon (Ne)-like Selenium plasma [11]. Almost at the same time Suckewer et al at Plasma Physics Laboratory, Princeton University (PPPL) demonstrated soft X-ray lasing at 18.2 nm wavelength from the Hydrogen (H)-like or Lithium (Li)-like ions. The two milestone discoveries were not only the first working soft X-ray lasers but also were the first working lasers in the x-ray spectral region.
1.6.2 Water Window X-ray Laser

The second x-ray regime is termed as water window, which is placed between the carbon and oxygen K-edges. The wavelength range is 4.4-2.2 nm. A strong X-ray laser was successfully demonstrated around 4 nm in 1990 by MacGowan at al. An X-ray laser at this wavelength will be perfect candidate for microscopy of living specimens in water.

1.6.3 Kilovolt X-ray Laser

The x-ray wavelength less than 1 nm is called kilovolt x-ray where photon energy is beyond 1 keV. The X-ray laser at this wavelength regime is very useful tool for high-precision holographic micro fabrication, non-destructive testing with phase-sensitive projection imaging [6]. Realizing X-ray lasers in the kilovolt x-ray spectral range is a big milestone and it is the main focused research area of this thesis.

1.6.4 Hard X-ray Laser

The fourth X-ray laser regime is the hard x-ray regime, which is more than 10 keV photon energy. Even though X-ray lasers in this regime has very attractive applications, to the knowledge of author of this thesis, only ideas have been proposed.

1.7 Available Approaches for Generation of Coherent X-ray Sources

Today there are various approaches generate coherent x-ray sources. The available approaches for generation of coherent x-ray sources is categorized as below in diagram 1.3 and briefly described in following sections.
1.7.1 Accelerator Based X-ray Sources

Accelerator based x-ray sources, synchrotron radiation and x-ray free electron lasers are currently producing extreme ultrabright x-rays. Following sections briefly discuss synchrotron radiation and x-ray free electron lasers.
1.7.1.1 Synchrotron Radiation X-ray Source

In synchrotron radiation, relativistic electrons are accelerated in a circular orbit and emit electromagnetic radiation in a broad spectral-range. The theoretical basis for synchrotron radiation traces back to 1897, Larmor derived an expression from classical electrodynamics for the instantaneous total power radiated by an accelerated charged particle. Although synchrotron radiation theoretically predicted by Liénard in the latter part of the 1800’s, the visible portion of this spectrum was first observed on April 24, 1947, at General Electric’s Schenectady facility by Floyd Haber, a machinist working with the synchrotron team [12]. The first synchrotron radiation was available for the application by mid 1970 [13].

Today one can use synchrotron-based crystallography to study the intricate details (at the atomic level) of very complex biological assemblies that have been formed into a crystalline sample. These developments have primarily utilized the high average brightness and broad spectral range of current-generation synchrotron x-ray sources.

The synchrotron x-ray sources from today’s typical third-generation x-ray sources have relatively long-pulse durations, ranging from tens to hundreds of picoseconds. Moreover, there are a relatively small number of coherent photons in the hard x-ray regime. These limitations result in two important scientific barriers. First, with present-day synchrotron x-ray sources, structure determinations with atomic or near-atomic resolution can only be performed on biomolecules (for example, proteins) that can be crystallized. Second, only static or very slowly evolving structures can be measured; however, the processes underlying biological function involve dynamically evolving molecular structures.
1.7.1.2 X-ray Free Electron Lasers

Free-electron lasers (FELs) are a laser that uses an electron beam from an accelerator to produce widely tunable, high power, ultrafast pulses of coherent radiation. John Madey at Stanford proposed the first idea for FEL in 1970 and John Madey and collaborators did first experimental demonstration of the FEL in 1976.

The concept of XFEL is almost similar to the traditional quantum lasers. A packet of about one billion electrons is sent through so-called undulator an arrangement of magnetic-fields with alternating field direction and accelerated. The accelerated charges particles emit synchrotron radiation at infrared or near UV wavelengths. A combination of theoretical, experimental, and technological advances has made it possible their extension to the x-ray region.

These extreme XFEL sources take advantage of true laser amplification to generate coherent beams that are exquisitely directed and focusable. Unlike current synchrotron x-ray sources, XFEL light will produce very short bursts of brilliant x-ray light with hundreds of femtosecond pulse-width. This shorter pulse-width is shorter than the movement of the atoms making up the biomolecule and eventually short, enough to capture the molecular structure before the molecule explodes as a result of the bright x-ray flash. The fourth-generation XFELs, such as the LCLS at SLAC, is to open up a completely new realm of x-ray science, enabling a new era of single bio-molecular and nanostructure determination as well as the ability to study structural dynamics in materials and chemical/biological systems. Detail parameters of currently available synchrotron and XFEL sources are presented below in table 1.1.
Today XFEL’s have achieved improved characteristics of peak-power, efficiency, coherence, higher repetition rates of operation, dramatically decreased pulse duration, and tune-ability. However, the use of accelerator-based ASE x-ray sources in a number of applications such as medical and industrials has been hampered by their size, complexity, and cost. This has set the goal of demonstrating laser action with enough brightness for potential application in an often-called tabletop compact X-ray laser that could be operated in university laboratory or hospitals. The coming section is describing such tabletop X-ray lasers.

<table>
<thead>
<tr>
<th>Facility</th>
<th>LCLS</th>
<th>BESSY</th>
<th>TESLA</th>
<th>MIT Bates</th>
<th>4GLS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Place</td>
<td>Stanford, United States</td>
<td>Berlin, Germany</td>
<td>Hamburg, Germany</td>
<td>Massachusetts, United state</td>
<td>Daresbury, United Kingdom</td>
</tr>
<tr>
<td>Type of source</td>
<td>XFEL</td>
<td>synchrotron</td>
<td>XFEL</td>
<td>XFEL</td>
<td>XFEL</td>
</tr>
<tr>
<td>Wavelength (nm)</td>
<td>0.15</td>
<td>51-1.24</td>
<td>6.4-0.1</td>
<td>100-0.3</td>
<td>124-12.4</td>
</tr>
<tr>
<td>Pulse duration (fs)</td>
<td>250</td>
<td>20</td>
<td>100</td>
<td>50</td>
<td>10</td>
</tr>
<tr>
<td>Peak Brightness (\text{photon/S.mm}^2\text{mrad}^2 0.1% BW)</td>
<td>(1 \times 10^{33})</td>
<td>(6.4 \times 10^{29} - 1.3 \times 10^{31})</td>
<td>((0.6-54) \times 10^{32})</td>
<td>(1 \times 10^{33})</td>
<td>(35 \times 10^{30})</td>
</tr>
<tr>
<td>Average Brightness (10^{33}), ((\text{photon/S.mm}^2\text{mrad}^2 0.1% BW))</td>
<td>2.7</td>
<td>Not available</td>
<td>30-1600</td>
<td>5</td>
<td>0.5</td>
</tr>
<tr>
<td>Peak power (GW)</td>
<td>8</td>
<td>1.5-14</td>
<td>135-24</td>
<td>4</td>
<td>Not available</td>
</tr>
<tr>
<td>Average power (mW)</td>
<td>250 @ 250 fs</td>
<td>120-200</td>
<td>((800-72) \times 10^3)</td>
<td>200</td>
<td>Not available</td>
</tr>
<tr>
<td>Number of photons per pulse (10^{12}) photon/pulse</td>
<td>1.1</td>
<td>0.2-70</td>
<td>430-1.2</td>
<td>0.3</td>
<td>70</td>
</tr>
<tr>
<td>Beam divergence (FWHM) (\mu\text{rad})</td>
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<td>37-140</td>
<td>27-0.8</td>
<td>Not available</td>
<td>Not available</td>
</tr>
<tr>
<td>Repetition rate (Hz)</td>
<td>30-60</td>
<td>1000</td>
<td>10</td>
<td>1000</td>
<td>1000</td>
</tr>
</tbody>
</table>

Table 1.1: Beam parameters of available synchrotron and XEFL sources [14], [15], and [16]. The parameters of LSLC are presented for its extreme wavelength at 0.15 nm.
1.8 Tabletop X-ray Lasers

Some of the most interesting and important applications discussed earlier are impossible to perform at centralized facilities such as synchrotron and XFEL. For example, studying proteins membrane and precious museum art objects are too fragile to be transported. Further, studying highly pathogenic proteins for which there is inadequate biohazard containment at the central facilities, and studying materials in the ultrahigh magnetic fields available only at specialized centers. In this sense compact-size tabletop coherence x-ray sources will be more available and user friendly than the larger machines.

The first working soft X-ray lasers invented in 1984 at LLNL and PPPL, are building-size large facilities they were almost quarter-size of current synchrotron facilities. The required pump-power for lasing action at X-ray laser made the soft X-ray laser at LLNL and PPL larger facilities. Realizing required power for x-ray lasing at the tabletop-scale was a dream until the CPA introduced to lasers in the middle of 1980. Today the CPA technique shrunk the building-size LLNL soft X-ray laser to three optical tables (5’x5’) tabletop X-ray laser

The first two working soft X-ray lasers were built based on traditional laser principle. There other x-ray lasers built based on nontraditional laser principles are also currently available. Brief descriptions of these ideas based on both traditional and non-traditional principles are presented below.
1.8.1 Laser Driven Plasma Based Tabletop Soft X-ray Lasers

Although the X-ray laser research pursued based on traditional laser principles, the field comes of age in 1984 with the first unambiguous demonstration of high-gain by groups at LLNL and PPPL. The both group used different approach to generate the gain. The LLNL used collisional excitation and PPPL used recombination excitation for creating population inversion in the gain-medium.

In the collisional excitation scheme, either Neon (Ne)-like or Nickel (Ni)-like ions are excited from the ground state (2p for Ne-like or 3d for Ni-like ions) to the laser upper-levels (3p or 4d, respectively). As the laser lower-levels (3s or 4p) are radiatively coupled to the ground state, they are depopulated by resonance emission and population inversion between 3p-3s or 4d-4p is created [17].

X-ray laser action in the recombination scheme requires hot plasma, that contains highly ionized bare nuclei or Helium (He)-like ions. Rapid cooling during the hydrodynamic expansion of the plasma causes fast recombination, that leads to the population of excited states of hydrogen (H)-like or Lithium (Li)-like ions. The first excited state is depopulated by resonance emission giving population inversion between the first and higher excites-states. Typical temperatures of plasmas to obtain population inversion of highly charged ions are several hundred eV for the collision excitation scheme and several tens of eV for the recombination one. In order to heat plasmas to such temperatures power density of laser radiation at the target should exceed $10^{13}$ W/cm$^2$ in the case of the collision scheme, and $10^{12}$ W/cm$^2$ in the case of the recombination scheme [17].
The first working soft X-ray laser in 1984 at the LLNL is the first laser driven plasma based soft X-ray lasers. Since their first demonstration, their size has been dramatically reduced, and their repetition rate and average power have greatly increased. Nowadays plasma-based soft X-ray lasers are compact, bright, tabletop device with unique characteristics that make them complementary to free-electron lasers and high harmonic sources. Their compact-size makes them ideal for installation in university and industrial laboratory settings.

In the LLNL the pump-laser is NOVA laser, which was used for inertial confinement fusion research. Typical parameter of the NOVA was wavelength 532 nm, 450 ps pulse-width, and 300 J energy. The NOVA was focused to a line-focus with focus dimensions 0.02 cm x 1.12 cm to generate $5 \times 10^{13}$ W/cm$^2$ intensity at the focal-plane on a solid 75 nm thick selenium solid-target, which is vapor deposited on one side of a 150 nm thick formvar substrate.

High-intensity line-focus of NOVA produces high-density and high-temperature plasma. In the Ne-like plasma, a large population of ions is collisionally excited to the 3p level. The 3s level has a relatively low population since it has a fast radiative transition to ground, and a population inversion is built-up between the 3p and 3s levels [10]. The population inversion in the atomic-levels $2p^5 3p$ and $2p^5 3s$ to $2s$ produces soft x-ray lasing at 20.63 and 20.96 nm wavelengths.

In 1987 at LLNL Matthews et al, first time, in the history of X-ray lasers, demonstrated the saturated amplification at which the total small signal gain was greater than ~ 16 and x-ray lasing wavelength was 20.6 nm. Presently at LLNL tabletop X-ray laser is configured differently from NOVA era. It uses compact multi-pulse terawatt
(COMET) laser driver to produce two pulses. First, a low-energy, nanosecond pulse of 5 J strikes a polished Palladium (Pd) or Titanium (Ti) target to produce the plasma and ionizes it. After the first-pulse, a second-pulse with 5 J energy and picoseCONDS **pulse-width** created by CPA technique arrives at the target a split second later to excite the ions and successfully generating Ni-like soft X-ray laser at 14.7 nm [18].

Immediately after the successful demonstration of soft X-ray laser at LLNL, another successful X-ray lasing demonstrated in PPPL used **recombination excitation**. A plasma column was created by irradiating a CO₂ laser with 75 ns pulse-width and 300 J pulse energy. In the **recombination** approach for H-like ions, the laser is used to create plasma with a high fraction of completely stripped ions. The plasma column was confined in strong solenoidal magnetic field of 9 Tesla (T) and cool it down by radiation losses. After the laser-pulse, the plasma is cooled rapidly and undergoes fast three-body recombination. The three-body recombination puts into upper excited levels a high-population, which decays downward by collisional radiative cascade and generate soft x-ray lasing. An observation of ~ 100 times enhancement of stimulated emission over spontaneous emission for the H-like carbon 18.2 nm radiation was the evidence for the soft x-ray lasing. Schematic for first recombination excitation soft X-ray laser at PPPL is presented in reference 19.

Further, research and development of the system, in 1990, made it possible to optimize the soft X-ray laser with output energy 1 to 3 mJ, 10 to 30 ns pulse duration, and a beam divergence of 5 milliradian at wavelength of 18.2 nm [10].

Since the realization of soft X-ray lasers at LLNL and PPPL, there have been so many different approaches and gain-mediumes were tested to realize possible shorter
wavelength X-ray laser and to reduce pump-energy. The challenging problem to improve the plasma based X-ray lasers is controlling the X-ray laser beam to propagate along the active-medium without any substantial refraction. To allow propagation along a gain direction without substantial refraction of the X-ray laser beam associated with transverse gradients in the electron density, the electron density distribution must be kept as uniform as possible in the active-medium. The plasmas produced by laser heating of solid-targets have typically large gradients, which result in severe refraction effects. Therefore, a number of methods for producing low-density gradients or for compensating the refraction have been applied in X-ray laser experiments. These methods includes laser exploding foils, magnetically confined laser plasma, double-target geometry, pre-pulse technique, and curved slab target. The last two methods have caused a significant improvement in the performance of a soft X-ray laser in terms of the peak-intensity and the beam divergence.

Since the success of x-ray lasing at LLNL and PPPL there are different methods proposed for realizing X-ray laser at possible shorter wavelength. One of the milestone development in the field of x-ray lasing took place in Max Plank Institute, Germany using a laser facility Asterix IV iodine. The approach is almost similar to the research presented in this thesis. In 1996 Fiedorowicz et al at the Max Plank Institute, Germany in collaboration with the group at the institute of Opto-electronics, Military University of Technology in Warsaw demonstrated strong evidence for Ni-like Xenon (Xe) (Z=54) lasing at 10 nm using a gas puff irradiated by a single picosecond laser-pulses. The Asterix IV iodine laser facility at the Max Plank Institute generates up to 600 J of light at 1315 nm in a 450 ps pulse-width. The laser output was focused to a 3 cm long by 150
mm wide line-focus by a 30 cm diameter cylinder lens array and focused on gas-flow through a 3 cm long and 400 μm wide sonic-nozzle. The experimental setup is shown in reference 20.

In the history of X-ray laser, in 1993, a breakthrough achieved by Da Silva at al at LLNL by demonstration of ultra-intense Yttrium (Z = 39) soft X-ray laser at a wavelength of 15.5 nm. The measured pulse-width was 200 ps and the measured peak X-ray laser power was 32 MW and the total output energy was 7 mJ which was the world’s powerful X-ray laser until the year 2002 [21].

1.8.2 High-orders Harmonic Generation

In High-orders Harmonic Generation (HHG), atoms exposed to a strong laser-field emit coherent light at frequencies much higher than that of the incident laser. Electrons can be ripped away from atom or molecule when the electric-field strength of an incident laser becomes comparable to the binding strength of electron to the atom. The intensities required $10^{13}$-$10^{15}$ W/cm² are easily accessible with tabletop femtosecond lasers. Once free, the electron will follow a trajectory controlled by the laser-field-first moving away from the parent ion, and then reversing in direction as the field oscillates. When it returns to the ion, it can possess significant amount of kinetic energy, much larger than the photon energy but being its multiple. This energy plus the ionization potential is transferred into emitted photon energy as soon as the electron recombines with its parent ion, which gives rise to very high harmonic orders with energy corresponding to dozens or hundreds of photons from the laser. Each time this happens, a burst of attosecond duration x-rays are emitted. This generally occurs twice during each
cycle of the driving laser field because the ionizing laser-field peaks twice each optical cycle. This high harmonic emission is **coherent** because each atom starts in the ground quantum state, and is exposed to the same **coherent** laser-field. Therefore, the evolution of the electron wave function that re-radiates the light is identical for each atom and adds coherently [22]. HHG is a result of the process of rescattering as described in the reference 22.

A classical picture of HHG gives a simple expression for the maximum energy photons that can be generated, which is simply the energy that the electron possesses at recollision is presented below by formula 1.6.

\[ h\lambda_{\text{max}} = I_p + 3.2U_p \propto I_L\lambda_p^2 \]  

(1.6)

Where \( I_p \) is the ionization potential, \( U_p \) is the ponderomotive potential, \( I_L \) is intensity of the pump-laser, \( \lambda_p \) is the wavelength of the pump-laser.

In the quantum physics aspects the electron during its trajectory as a “free” particle, evolves with a de-Broglie wavelength (\( \lambda_p \)) corresponding to the formula 1.7 presented below.

\[ \lambda_p = \frac{h}{p} \]  

(1.7)

Where \( h \) is Plank constant, and \( p \) is momentum.

If one calculates the total phase advance of the electron during its free trajectory, it is quite large—dozens to hundreds of radians. The optical phase of the emitted harmonic light depends on this electron’s phase, and thus is not rigidly related to the phase of the driving laser. It depends on the exact shape of the electromagnetic field of the laser during the sub-optical-cycle time of the rescattering trajectory [22]. Bright harmonic beams are emitted if the emissions from many atoms in a medium are combined together.
coherently. The mechanism is called Quasi-Phase-Matching (QPM) techniques. The high-harmonic spectrum and pulse duration can be controlled by manipulating the macroscopic phase matching conditions and the electron recollision process. This allows the pulse duration to be adjusted from tens of femtoseconds to less than 100 attoseconds—the shortest pulse durations of any light source to date. One great advantage of HHG sources is that the emission is perfectly synchronized to the driving laser. To date, bright high harmonics at photon energies of ~ 0.5 keV has been achieved and maximum energy per pulse achieved in the XUV wavelength is less than 5 µJ. The parameter of available HHG x-ray sources is presented at table 1.2.

1.8.3 Inverse Compton Scattering

Mechanism of Inverse Compton Scattering (ICS) is the scattering of electrons on photons, which generates x-ray radiations. From a classical point of view, the process is referred to as “Thomson scattering” and can be visualized as a plane-wave of frequency $\omega_0$ impinging on an electron. In the approximation where laser energy is much lower than rest electron energy in the rest electron frame, the scattered electromagnetic wave frequency is conserved. In the laboratory frame, as electrons are relativistic, the incident electromagnetic wave undergoes two frame changes, and its frequency is increased by a $4\gamma^2$ factor due to the relativistic Doppler effect. In quantum electrodynamics, ICS is visualized as a succession of absorption and emission of a photon by an electron. In this approach, it is referred as “Compton scattering,” which consider electron recoil and particle polarization. The process is described as the collision between photons and electrons. This collision is elastic, so the interaction parameters can be calculated
according to energy and momentum conservation. The scattered photon energy is a function of electron energy, laser wavelength, collision, and scattering angles. The concepts of Compton scattering are shown in the figure 1.2 below.

![Figure 1.2: Schematic view of photon electron interaction in Compton scattering](image)

Since the ICS sources are compact, it will be an ideal source for the places where unique science can be done in university nano-centers or pharmaceutical companies. Nowadays the commercial ICS sources are available in market. A typical commercial ICS source is capable of generating flux of $10^{11}$ photons/s/0.1% BW and average brightness of $10^{12}$ photons/s/mm$^2$/mrad$^2$/0.1% BW is suitable source for crystallography and phase-contrast imaging. A ICS source at Brookhaven National Laboratory (BNL) accelerator test facility has demonstrated peak brightness in excess of $10^{20}$ photons/s/mm$^2$/mrad$^2$/0.1%BW with $5 \times 10^8$ photons/pulse at 10 Hz in a 50% bandwidth [14].

1.8.4 Miscellaneous X-ray Sources

In addition to the methods mentioned above there exist different approaches to generate tabletop coherent x-ray sources. In 1993 at Colorado State University, Rocca et al realized tabletop Ne-like soft x-ray lasing at 46.9 nm using discharged pumped fashion [23]. A fast capillary discharge having a 10-90% current rise time of 20 ns, 40 kA was
used to excite plasma columns up to 12 cm in length in 4 mm channels, producing population inversion. After further research and development Colorado State University demonstrated (gain-length > 25) saturated amplification soft X-ray laser at 46.9 nm with 30 μJ energy, and 7 Hz operation [6].

Another method of producing directional x-ray sources is Larmor radiation excitation emitters. In 1999, at advance photon research center Ueshima et al proposed Larmor radiation to generate ultra-short intense directional x-ray beam. In, ponderomotively-generated, Larmor radiations excited emitter, ultra-short high intensity laser irradiate on relativistic plasma in which the free electrons are forced to move in certain way, resulting in the production of an ultra-short intense directional x-ray beam with narrow spectral band.

The parameters of currently available x-ray sources are presented in table1.2 and figure 1.3.
### Table 1.2: Parameters of currently available x-ray sources [14].

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Tabletop soft X-ray lasers (Seeded)</th>
<th>HHG (1-W driving laser 100 mj, 10 Hz)</th>
<th>Tabletop capillary discharged laser</th>
<th>FLASH LLNL, USA</th>
</tr>
</thead>
<tbody>
<tr>
<td>Photon energy</td>
<td>25-94 eV 49.65-13.2 nm</td>
<td>20-500 eV 62-2.48 nm</td>
<td>26.5 eV 46.84 nm</td>
<td>10-200 eV 124-6.21 nm</td>
</tr>
<tr>
<td>Energy/pulse</td>
<td>0.01-0.1 μJ 1 μJ @ 50 eV 0.1-0.5 mJ</td>
<td>10-100 μJ</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Repetition rate</td>
<td>1-10 Hz 10 Hz 3-10 Hz 1-10 Hz</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Pulse duration</strong></td>
<td>1 ps 10 attosec-30 fs 1 ns</td>
<td>1 ps</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Focused intensity</td>
<td>~10(^{14}) W/cm(^2) ~ 10(^{15}) W/cm(^2) ~ 4x10(^{13}) W/cm(^2) ~ 4x10(^{14}) W/cm(^2)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Average power</td>
<td>0.1-0.5 μW 10 μW 0.1-2 mW 0.1-0.5 μW</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Coherence</td>
<td>Full spatial and temporal</td>
<td>Full spatial and temporal</td>
<td>Full spatial</td>
<td>Full spatial and full temporal</td>
</tr>
<tr>
<td>Polarization</td>
<td>Linear</td>
<td>Linear</td>
<td>Un polarized</td>
<td>Linear</td>
</tr>
</tbody>
</table>

Figure 1.3: Brightness of currently available x-ray sources [6].
The table 1.2 clearly shows that the average output-energy of the currently available x-ray sources are in the order of micro-joule and none of the currently available x-ray sources are operating at the wavelength less than two nanometers. The figure 1.3 shows that the none of the currently available x-ray sources successfully generating coherent radiation in excess of $10^{30}$ photons/sec/mrad$^2$/mm$^2$ (0.1 % bandwidth) at a wavelength less than one nanometer, The brightness level $10^{30}$ photons/sec/mrad$^2$/mm$^2$ (0.1 %Band width) is sufficient for interferometric biological imaging capable of providing an atomic-level resolution visualization of the molecular anatomy of cells, tissues and organisms in the natural state [24]. The spatial-resolution at different wavelength, presented in appendix 1, clearly shows that the x-ray sources with less than 2 nm operating wavelength are suitable candidate to explore the atomic world.

It is claimed that an apparatus and method at the XRIM, UIC is success in generating ultrabright keV x-rays from saturated amplification on noble gas transition arrays from hollow-atom states [24]. An estimate of the peak brightness achieved in the experiments gave a value of $\sim 10^{30}$ photons/sec/mrad$^2$/mm$^2$ (0.1 % bandwidth) at the wavelength less than 1 nm [25]. The operating wavelength and brightness of the x-ray source make it as a unique extreme x-ray source over currently available tabletop x-ray sources to date.

1.9 X-ray Micro Imaging and Bioinformatics Laboratory

The x-ray micro imaging and bioinformatics laboratory team is in the research field of generation of ultrabright x-ray source for more than 30 years. The tirelessly
experiments and theoretical works performed at XRIM sought fruitful solutions for the challenges of generation of coherent source in the x-ray wavelengths less than one nanometers regime.

As mentioned earlier in the section 1.5, one of the major challenges for realizing tabletop coherent x-ray source is required higher pump-power for generating population inversion and maintaining populating inversion. XRIM team resolved the problem by using a completely new mode of controlled concentration of powers, which involves the combination of ultrahigh brightness of femtosecond laser technology, multi-photon coupling to atoms and molecules, and a new mode of electromagnetic propagation [25]. Detail descriptions of technology of XRIM are presented in chapter 3.

The typical experimental setup in the early stage of the research at XRIM is shown below in diagram 1.4 [25].

Diagram 1.4: Previous experimental setup at XRIM [25]
In the previous experiments, a 1.5 mm diameter sonic-nozzle sprays Xe gas inside evacuated experimental-chamber. Target-clusters are formed in the gas jet due to the adiabatic cooling as presented below. The pressure difference between the sonic-nozzle stagnation pressure (150 psig) and the target-chamber (typically at $10^{-4}$ mbar) results the target-gas adiabatic expansion into vacuum and cooling it down. The low-temperature target-gas results the Van der Waals forces between atomic become dominant and forming target-cluster. An off-axis parabolic mirror focuses the KrF* UV laser on target-cluster just right after it formed. Produced pump-laser intensity at the target shows that 1 watts per atom is possible [26]. The physics, which takes place in the experiments, follow in the next paragraph.

The fundamental idea of generation of radiation at x-ray wavelength at XRIM is removing inner-shell orbital electrons from high Z Xe/Kr target-clusters and creating species of hollow-atom states. Series of experiments and theoretical works performed at XRIM confirmed that the direct selective inner-shell excitation is possible [27], and [28]. The inverted populated hollow-atom species emits x-ray radiation at specific wavelengths. The theoretical research works further proved that individual multi-photon clusters can exhibit superradiance in the 1.24-0.12 nm (1-10 keV) spectral-range [29]. The superradiance process may (i) confer a high-level of coherence on the emission of an x-ray amplifier (ii) provide control of the emitted wavelength, and (iii) produces very short x-ray pulses. The action of supper radian results coherence emission of ultra bright x-ray radiation at certain x-ray wavelength with possible short pulse-width.

A physical process, termed in this thesis as self-channeling, confines the ultrabright x-ray radiation and channeling it as a directional x-ray source. The
experimental and theoretical works performed at XRIM on high-intensity short-pulse propagation in plasmas confirms evidences for the formation of stable mode of spatially confined channel propagation [30], and [31].

The concept pyramids, presented below in diagram 1.5, built based on theoretical and experimental research works performed in the XRIM shows road to success in generating ultrabright x-ray source in the wavelength region less than 2 nm.

Diagram 1.5: XRIM concept pyramid to succeed tabletop keV ultrabright x-ray source.
Three types of x-ray sources are realized and presented in thesis. According to the x-ray source wavelength range and type of gain-medium involved the sources, they are named as described below.

1. Xe L-shell x-ray radiation: Wavelength 0.25 – 0.33 nm (5.2 – 3.8 keV)
2. Xe M-shell radiation: wavelength 1.1 – 1.6 nm (1127 – 775 eV)
3. Kr L-shell radiation: wavelength 0.55 – 0.80 nm (2500 – 1550 eV)

The above presented discussions clearly show that generation of tabletop ultrabright x-rays source in the wavelength less than 2 nm will be a complementary x-ray source to the XFEL and synchrotron x-ray radiation facilities. To date any other research groups, succeeded in generating such x-ray source in laboratory-scale.

Since the previous experiments performed in XIRM clearly showed evidences for the realization of tabletop coherent L-shell x-ray source, the XIRM team was motivated to optimize and characterize the realized unique and extreme tabletop x-ray source. Research works focused to optimize the realized x-ray source via upgrading laboratory, pump-laser system, diagnostics, target-medium, and all other experimental parameters related to the x-ray source. This thesis is mainly focusing generation, characterization, and optimization of tabletop ultrabright directed Xe M-shell, and Kr L-shell keV x-ray radiation sources. The size of the realized x-ray source is 3 optical-tables (5’ x 12’) plus two KrF* amplifiers. The following chapter 2, experimental methods and apparatus, describe the detail methods and apparatus for generation and characterization of the tabletop ultrabright Xe M-shell, and Kr L-shell x-ray sources.

😊 ————————— END OF CHAPTER 1 ————————— 😊
2. EXPERIMENTAL METHOD AND APPARATUS

2.1 Introduction

This chapter is a detailed description of the experimental method and apparatus for the development of tabletop ultrabright keV x-ray sources from Xe and Kr hollow-atom states at the X-Ray Micro-Imaging and bioinformatics laboratory (XRIM).

In first section of chapter 2, experimental-method begins with the fundamental steps needed for generation of tabletop-sized ultrabright directional x-ray sources, while theory and physical processes behind each of these steps are presented, briefly, in chapter 3. At the end of the first section, experimental-methods, a list of apparatus needed for the success of the experimental-methods is presented.

Second section, apparatus begins with discussing ideas for improving the x-ray source-quality and then it describes the design, the engineering, the operations, and the alignment procedure needed for the diagnostics and for successful optimization and characterization of the x-ray source of the XRIM.

2.2 Experimental Method

The experimental-method need to generate tabletop-sized ultrabright directional Xe \textit{M-shell}, and Kr \textit{L-shell} x-ray sources, at XRIM, are simple, effective and unique. The steps presented below briefly explain the experimental-method used to generate an ultrabright directional Xe \textit{M-shell}, and Kr \textit{L-shell} x-ray sources, as well as its optimization and characterization.
Step 1: Cluster formation

The first-step is the target atomic-clusters formation using a solenoid-poppet sonic-nozzle. The solenoid-poppet sonic-nozzle is termed, in this thesis, as Double Gas Sonic-Nozzle (DGSN). A custom designed DGSN opens and then sprays target-gas inside the experimental-vacuum-chamber few milliseconds before the pump-KrF\(^+\)-laser interacts with target-cluster molecules. The adiabatic expansion of the target-gas inside vacuum-chamber results to cooling down the target-gas. The cooling allows the Van der Waals force to dominate and strong-enough to bring the gas-atoms to form target-cluster molecules. Section 3.3 of chapter 3 presents physics behind the target-cluster formation. The section 2.3.2 of the chapter 2 presents the design of the DGSN and the mechanical details of the sonic-nozzle. The Xe and Kr clusters are the main target-material in XRIM.

Step 2: Excitation

The second-step in a realization of laboratory sized ultrabright directional x-ray sources is the exciting and the creation of hollow Xe/Kr atom states (lacking an inner-shell with outer shell-electrons presented). In the XRIM, the excitation mechanism is triggered by KrF\(^+\) pump-laser. The KrF\(^+\) beam is focused to diffraction-limited (defined in the section 2.3.1.2.1) focal-spot and then it interacts with the target-clusters molecules. A timing device, Stanford box (model No DG 535), perfectly synchronizes the diffraction-limited pump-laser focused-spot to arrive when the target-clusters have formed. The techniques and procedures for focusing the KrF\(^+\)-laser beam to diffraction-limited focal-spot size and align the focused-spot to target-clusters are presented below in
the section 2.3.1.2 of apparatus. Section 3.4 of chapter 3 presents the detail physics and theory of the excitation mechanisms.

Step 3: Channel formation

Channeling generated x-ray radiations in the forward axial-direction is the third-step. The femtoseconds UV pump-laser pulses not only contribute to creating of hollow atom states, but also channel the x-ray radiation to form directional x-ray source. Section 4.2.2.1 of chapter 4 presents the experimental evidences for the channel-formation and section 3.5 of chapter 3 presents a developed theory to understand the channel-formation.

Step 4: Monitoring and optimizing

In the experiments presented in this thesis, two von Hámos single-crystal spectrometer (one is in the axial-direction and other one is in the transverse-direction), a triplet-aperture x-ray camera, Thomson scattering optical-setup, and a calibrated fast x-ray photo-diode are visualizing target-area and recording single-shot data every time pump-laser interacts with target-clusters. The temperature and the backing pressure of the target-gas, and UV pump-energy are also recorded every time pump-laser interacts with target-clusters. The section 2.3, experimental-apparatus, presents detailed designs, and engineering descriptions of the diagnostics. The experimental single-shot outputs were monitored real-time and recorded during the experiment for each shot.

In a typical experiment, the experimental-parameters such as target-gas pulsed jet stagnation pressure, target-gas temperature, the timing between the UV pump-laser pulse and the DGSN solenoid activation as well as DGSN spatial position relative to UV focus
were changed and the effects of the changes in x-ray signals are monitored in real-time and recorded. During the experiments, the experimental-parameters were monitored in real-time and tuned them such a way to optimize the realized x-ray source. In order to characterize, understand, and optimize the x-ray source the axial-von Hámos single-crystal spectrometer moved small steps along the on-axial direction of propagating of the pump-laser and the spectra at different spatial-locations was recorded. Further, the Bragg angle of the axial-von Hámos spectrometer was changed and the corresponding spectrum recorded. The chapter 4, results and discussions, presents details of experimental results along with explanations.

2.3 Experimental Apparatus

X-ray micro-imaging and bioinformatics laboratory pioneers in the field of generation of laboratory size ultrabright x-ray source at wavelengths less than 2 nm. In the middle of 2008, there were series of experiments performed at the XRIM laboratory to realize a laboratory-size X-ray laser at wavelengths less than 2 nm. The experimental results of the realized x-ray source showed convincing evidences for X-ray laser-like properties [25] at the wavelength less than 2 nm. This x-ray source required a quality study in order to use it for potential applications presented in the chapter 1. The diagnostic instruments used in that experiment are designed in 1990. The obsolete and outdated 1990’s diagnostic instruments are not capable enough to characterize the x-ray source completely. A complete characterization of x-ray source demands total diagnostic upgrade to use latest technologies.
The diagnostic upgrade and design requires understanding experimental parameters to optimize and characterize the x-ray source. The previous experimental and theoretical works performed in the XRIM clearly showed that the optimization mainly depends on the following parameters.

I. Brightness of the pump-laser, and spatial pointing-stability of focused-spot on clusters-target

The unwritten fundamental rule in the filed of X-ray lasers is that the more pump-energy supplied to the target-clusters, the more it radiate back as x-rays. In order to deliver possible higher pump-power at the target-clusters, one should first optimize the brightness of the pump-laser. The brightness of the pump-laser beam depends on focal-spot size at the target, pulse-width of the pump-laser, spatial and temporal-profile of the pump-laser-beam.

Delivering the diffraction-limited focused beam on the target-clusters in a controllable fashion is the key parameter to control ionization mechanisms that will lead to an optimized x-ray source. Further, the pump-laser not only creates hollow Xe / Kr atom states but also supports channeling process for the x-ray radiation. Therefore, launching a higher brightness diffraction-limited focused-beam on target-clusters with a correct timing under a controllable method is one of the key to optimize the ultrabright x-ray sources quality.

In the XRIM, an Off-Axis Parabolic mirror (OAP) is the optic, which focuses the pump-laser-beam to a diffraction-limited focal-spot, and proper alignments can produce the necessary pump-power for the generation of ultrabright x-ray source at wavelengths
less than two nm. To produce a diffraction-limited focal-spot the OAP is mounted on a custom-designed six-axis positioner. An optical-setup termed in this thesis as “Focal-Spot-Monitoring Optical-Setup” (FMSOS) is designed to produce a real-relay-image of the magnified single-shot focused-spot on a Charge-Coupled Device (CCD) camera chip. The FMSOS consists a micro-spot focusing-objective and a CCD camera both mounted separately on five-axis positioners. The section 2.3.1.2.2 presents detailed designs of both devices and alignment procedures.

The optimization of the brightness of the pump-laser mainly depends on the pump-laser-system and focusing-devices. Diagram 2.1 below shows, at a glance, how the instruments and apparatus influence the brightness of a pump-laser.

![Diagram 2.1: Apparatus for optimizing brightness of the pump-laser.](image)

Like the brightness of the pump-laser, the spatial-pointing stability of focused-spot on target-clusters also decides quality of the generated x-ray source. A higher beam-pointing stability for the focused-spot will result the higher beam-pointing stability of the
generated x-ray source. Further, higher beam pointing-stability will ensure reproducibility to the x-ray source in experimental-data.

The following apparatus, presented below, are play important roles to optimize and the beam-pointing stability of the pump-laser on target-cluster.

i. Optical-mounts and holders for beam-steering mirrors.

ii. Optical-tables for laser-system and experimental-chamber.

iii. Vacuum-system though which pump-laser-beam propagates.

iv. Experimental-chamber.

The design and alignment procedure of the above apparatus are discussed in the later part of this chapter 2.

II. Density and size of target-clusters

DGSN produces the Xe/Kr excitation medium for x-ray generation. Cluster-molecular size, shape, and density are important factor, which decides the inner-shell ionization mechanism. The shape and size of the sonic-nozzle orifice, target-gas backing-pressure, target-gas temperature, and experimental-chamber vacuum level are some of parameters determine the target-clusters size, shape, and density. The section 3.3 of chapter 3 presents the physics behind the target-cluster formation.

The plasma formed during the laser-matter interaction consists of target-ions, electron, and the unionized target-gas molecules. Physical mechanism involved during the generation of ultrabright x-ray sources is an ultrashort event and the unionized target-gas molecules moves at the speed of sound after ejected from the DGSN. Therefore, the unionized target-gas molecules stay in the vicinity of generated x-ray source during the generation of ultrabright x-ray source and the clouds of unionized target-gas-molecules
will attenuate propagating x-ray beam. Therefore, confining the target-gas molecules into a smaller volume with definite boundaries might reduce the x-ray attenuation by the unionized target-gas molecules. Further, it is believed that once the directional x-ray beam emerges from this smaller volume, where the unionized target-gas molecules are trapped, then a directed x-ray beam will propagate without attenuation for a long distance.

The amount of attenuation when x-rays pass through low-Z molecules (such as nitrogen (N) or helium (He)) is considerably smaller than compared to the amount of attenuation when the x-ray beams travel through the high-Z target-gas molecules (such as Xe or Kr). Therefore, it was proposed to shield the target-gas molecules by low-Z molecules during the generation of ultrabright x-ray sources. DGSN is designed for this purpose. In the design, the primary target-gas molecules will be sprayed in the center of the nozzle while low-Z secondary gas-molecules are sprayed within an annular area of the target-gas. The annular low-Z gas-molecules with the proper conditions for backing pressures of both gases, and the opening times of both gases poppet valves compress the target-gas molecules into a small volume and form definite barriers to target-gas-molecules. The section 2.3.2 presents the mechanical designs of the DGSN along with the experimental results.

The generations of directional and ultrabright x-ray sources experiments are conducted inside a vacuum-chamber. Since the proper spatial-alignment of the target-clusters to the pump-laser focused-spot will lead to optimize the laser-matter interaction and result in an optimized x-ray source. The DGSN is mounted on a three-axis positioner
to align the target-clusters at the pump-laser focused-spot during the experiments. The section 2.3.2 presents details design of the DGSN positioner and, alignment procedure.

The apparatus play important roles for optimizing x-ray source via tuning the density, shape, and size of the target-clusters and spatial-alignment of the target-clusters to the pump-laser are summarized below.

i. Double-gas sonic-nozzle (DGSN): Formation of target-clusters and confining the target-gas into smaller volume.

ii. Three-axis positioner of sonic-nozzle: Optimize spatial-alignment of the target-clusters to the pump-laser focal-spot.

iii. Alignment tools: Helps to align the pump-laser focal-spot at the center of DGSN orifice and helps to align diagnostics in their optimized alignments.

III. Experimental-diagnostics

The optimization of the x-ray source demands knowledge of mechanism involved during the laser-matter interaction. Reliable diagnostics are ones that monitor the x-ray source and collect detail information of what takes place during the experiments. The experiments are conducted inside a vacuum chamber. Once the vacuum chamber is pumped-down, the diagnostics setups provide visualization into the vacuum-chamber and monitor experimental-parameters to guide optimization of the generated x-rays source. Therefore, the diagnostic setups are not only a tool to record the data, but also give real-time single-shot information inside the experimental-chamber during the experiments.

In the previous experiments, target-area was monitored with only an aperture x-ray camera and a spectrometer. Optimizing number of diagnostics within the target-area will provide many details about the physics take place. To optimize number of
diagnostics and to align all the diagnostics to their optimized alignment without interfering with any other diagnostics, demands a careful design and engineering. In the experiments, presented in this thesis, the number of diagnostics monitoring the target-area is significantly increased compare to the previous experiments. There are at least seven different calibrated diagnostics viewing the target-area with ten different experimental-single-shot parameters recorded every time the pump-laser interacts with the target-cluster molecules.

Alignments of all these diagnostics are performed prior to begin the experimental-chamber pump down to high vacuum. It is inevitable that the vacuum pumping-down process will misalign the diagnostic alignments. Therefore, it is important to make compensation corrections for any possible misalignment of diagnostics caused by evacuating the experimental-chamber. Careful design and engineering enables the diagnostics to be aligned inside the vacuum chamber, during the experiments by viewing single-shot data in real-time.

Aligning all the diagnostics, every day, to same modus operandi, guarantees reproducibility to experimental-data sets. Further, the collected data sets at different experimental-date will be comparable, only if the diagnostics are aligned in same modus operandi. However, alignment of all diagnostic everyday to the same precision is a major challenge to diagnostic design of the X-ray laser experiments. Further, aligning all the diagnostics around a compact target-area in a reasonable-time without interfering with other diagnostics presents another major challenge in the diagnostic design and engineering of the X-ray laser projects.
Diagnostics infrastructure around the target-area of the experiments, presented in this thesis, is carefully designed to keep all diagnostics in permanent locations to view target-area at their best alignment positions. The design is avoiding removal of diagnostics during the day-today alignment. The diagnostics requires, prior to begin experiments, only a simple alignment check to make sure they are in their best alignment position. The diagnostics design and diagnostics infrastructure design not only save time but also guarantee reproducible data sets. The details designs and engineering of each diagnostics are presented in the section 2.3.3 and alignment procedures are also presented in the same section followed by the design. Figures 2.17, 2.18, and 2.19 show the diagnostics infrastructure.

The list of apparatus for monitoring and recording the experimental-parameters are presented below

i. Axial-von Hámos single-crystal spectrometer.

ii. Transverse-von Hámos single-crystal spectrometer.

iii. Triplet-aperture x-ray camera, which is capable of imaging simultaneously two different x-ray wavelength radiations from same x-ray source.

iv. Thomson scattering setup to visualize scattered pump-laser UV light from free-electrons.

v. Calibrated fast x-ray photo-diode to measure x-ray energy.

vi. Thermocouple and pressure-gauge to record target-cluster temperature and backing-pressure respectively.

IV. Analyzing experimental-results

Analyzing experimental-results in a creative fashion is an important key to understand the mechanism involving in the experiments, which will lead to optimize the x-ray source. The chapter 4 discusses the details of analyzing the experimental-data and the analyzed results are also presented in the same chapter.

The following software is used to analyzed data and in designing experiments.

i. AutoCAD; Computer-aided design (CAD) and drafting by Autodesk, version 2011.

ii. SolidWorks; CAD, drawing design and 3-D modeling by Dassault systèmes solidworks corporation, version 2012

iii. IDL; Programming and visualization software by Exelis, version 8.2.1.

iv. MATLAB; Numerical computing environment and programming language by MathWorks, version R2012b.

v. Lab VIEW; Graphical programming environment by national instruments, version 2012.

Diagram 2.2 presented below is summarizing, at a glance, all the x-ray source optimizing parameters.
Diagram 2.2: X-ray source optimizing parameters at a glance
In this thesis, apparatus design is presented, according to the above diagram 2.2, along with alignment procedure.

2.3.1 Apparatus Design to Optimize Pump-laser Brightness and Beam Pointing.

Optimizing brightness and beam-pointing of the pump-laser is the fundamental candidate to optimize the ultrabright x-ray source. As presented in the above diagrams 2.1 and 2.2 the brightness of the pump-laser is depend on the laser-system and focusing-devices. The following section, laser-system, discusses, laser-system design, beam-line design, experimental-chamber design, and beam-alignment procedure.

2.3.1.1 Laser-System

The entire laboratory along with the laser-system is redesigned for the research experiments presented in this thesis. In the 30 years history of XRIM, there are various laser-systems built to study generation of ultrabright x-ray source. Since history is a mentor, which channels the future towards the right path, first of all previous laser-systems are studied to understand pros and cons of these laser-systems. The advantages of the old laser-systems are implemented to the new laser-system while the disadvantages of the old laser-systems are redesigned.

In the 30 years history of XRIM, there have been five laser-system built to study generation of ultrabright x-ray source. The laser-systems are termed, in this thesis, as first, second, third, fourth and fifth generation laser-systems. The details of first three laser-systems are not presented in this thesis. However, the details of first three laser-systems are presented in references 34, 35, 36, 37, and 38 respectively. The discussion of
laser-system begins with the fourth-generation laser-system as the research presented in thesis was started in the laser-system.

2.3.1.1 Fourth-Generation Laser System

The experimental research presented in this thesis born in this laser-system and accomplished in the next generation laser-system. In the winter of 2005, while the custom designed third-generation laser-system have been using for experiments, a new compact-size fourth-generation laser-system was built from commercial products.

The fourth-generation pump-laser for generation of x-ray source at XRIM begins with the commercial spectra-physics seeding laser-system. The spectra-physics product, Hurricane, is a regenerative-amplifier system including all the solid-state technology in a compact 48" x 36" x 10". It comprises a seeding-laser (MaiTai), a pump-laser (Evolution), a stretcher, a Regenerative Amplifier (RA), and a compressor. The typical output of the Hurricane laser has 50 μJ energy, 120 fs pulse-width, 745.8 nm wavelength, and 8 mm circular Gaussian beam. The schematic diagram of the Hurricane is presented in the appendix 2.1.

The output of the Hurricane is sent to spectra-physics tripling unit where the Hurricane output beam is tripled up to 248.6 nm wavelength for the KrF* excimer-amplification. The schematic diagram of the tripling unit is also presented in the appendix 2.1. In the tripling unit, the input fundamental first generates the second harmonic in SHG crystals and then the third-harmonic $\omega_3$ is generated by the frequency summation of the remaining fundamental $\omega_1$ and the second harmonic $\omega_2$ in a THG crystal.
The tripler output-beam is expanded and focused to a small spot-size inside vacuum. The diverging beam from the focused-spot is collimated and then sent to a KrF\textsuperscript{*} excimer-amplifier, name LLG TWIN AMP, in off-axis fashion for the first two-stages of amplification. The second-stage pre-amplified output from LLG TWIN AMP sent to double-grating compressor-pair. The compressed-beam from the output of grating-pair sent to vacuum ASE spatial-filter. The spatial-filtered beam after expansion and collimation is sent to the second tube of LLG TWIN AMP in off-axis fashion for third and then final fourth-amplification. The final pre-amplified beam is sent to a Beam-Expanding-Telescope (BET), which is a replacement of the previous beam expander Dall-Kirkham telescope. The schematic diagram of the laser-system is presented in the appendix 2.2. The output of BET is sent to the final KrF\textsuperscript{*} amplifier known as Prometheus. Details of Prometheus are presented below in the following paragraph.

The Prometheus KrF\textsuperscript{*} amplifier added to the second-generation laser-system. Prometheus is one of the two identical amplifiers, one delivered to Los Alamos National Laboratory (LANL) and the other to XRIM in the spring of 1988 by Beta Development Corporation. The main differences between both machines are different gain-medium and different operating pressure. The gain-medium of the Prometheus is KrF\textsuperscript{*} and the operating pressure is 1.75 mbar while the gain-medium of the LANL was XeCl and the operating pressure at 2 bars. The XRIM Prometheus is a unique machine as LANL terminated operation of their machine because of its poor operation.

Prometheus was specifically designed to work at relatively low-pressure and low-gain in order to reduce wave-front distortion and Amplified Spontaneous Emission (ASE), conditions that are necessary for the preamplifier beam quality. Prometheus has
two parallel 2.5 m long discharge-channel separated by an x-ray gun preionizer. Although the system has capability to excite a discharge with a total cross-sectional area of 10 cm x 10 cm, the present device, owing to excessive curvature of the electrodes, produces a saturated output only over approximately one third of that area. Typical operating conditions employ an x-ray-gun voltage of ~115 kV, a discharge voltage of ~180 kV, a pulse repetition rate of 0.1 Hz, and a 1.6 atm gas mix of 0.15% Fluorine (F₂), 5% Kr, and 94.85% He. Measurements give a small-signal gain of ~2.9% cm⁻¹ and a non-saturable loss parameter of 0.28% cm⁻¹.

The gas flow between the discharge electrodes is laminar and transverse with a flow rate sufficient to clear the discharge volume nearly 5 times between laser pulses. The laser medium and the electrical insulating oil used in the high-voltage enclosure are temperature controlled to minimize thermal gradients and consequent wave-front distortion in the optical-path. A measurement of beam-distortion of 632 nm with full gas flow, but without the discharge operating, demonstrated an RMS wave-front distortion of λ/13 over the full aperture. Further, measurements showed that the contribution to nonlinear distortion from the amplifier medium, mainly helium, is negligible. Under these conditions, the spatial character of the output beam is governed by that of the seed beam [37].

During its first operation, output energy of ~250 mJ was produced, with an input pulse of ~200 µJ. The associated ASE, which mainly originates from the power amplifier itself, is small and poorly focusable. The ASE comprises ~10% of the short pulse energy and has a measured minimum focal-diameter of ~3 mm [37]. It was concluded from the measurement that the Prometheus along with 2nd generation laser-system was capable of
achieving average intensity of $\sim 2 \times 10^{19}$ W/cm$^2$, which is corresponds to peak electric field of $\sim 24(e/a_0^2) = 1.2 \times 10^{11}$ V/cm exceeds the characteristic of the outer electronic shells in all materials.

In the 30 years history of XRIM laboratory, the laser has faced so many changes and modifications from the day Prometheus installed up to today nothing could replace the heart of the laboratory; the Prometheus. The reliable operation and uniqueness of Prometheus makes the XRIM as the one of the world-class high intensity KrF*UV laser-system.

The final output of Prometheus sent through vacuum-tubes to an experimental-chamber (termed in this thesis as small chamber) built in the far North-West corner of XRIM during the third-generation laser-system. The size of the system from oscillator to small chamber is 11 optical-tables (6’x12’x1’.) plus LLG TWIN AMP and Prometheus. The typical laser-parameters inside chamber were $\sim$ 300 mJ average energy, 500 fs pulse-width, and 9 x 8 cm beam-size. The maximum intensity produced by the laser-system was $10^{20}$ W/cm$^2$, which is 10 times higher than the previous third-generation laser-system.

The experiments performed using the laser-system and the small chamber clearly showed the realized x-ray source has strong evidences for lasing actions at the 0.29 nm wavelength. Since an ultrabright laboratory size X-ray laser at 0.29 nm wavelengths will be a revolutionary source for various applications the XRIM team was motivated to optimize and characterize the realized x-ray source. Completely new laser-system built for the purpose of reproducing the x-ray source again and then optimizing it. The following section, fifth-generation laser-system, is discussing the design and engineering
of the fifth-generation laser-system. The fifth-generation laser is the pump-laser for the experiments presented in this thesis.

2.3.1.1.2 Fifth Generation Laser System

The fifth-generation laser-system is built for optimizing pump-power and beam stability at the target-clusters to provide ultimate pump-power with controlled fashion. First of all, detail research was performed to find out the pros and cons of the previous fourth-generation laser-systems. A diagnostics setup was built to study the beam-quality and pump-laser beam pointing-stability. The built diagnostic-setup is exactly similar to the small-chamber. The diagnostics beam was focused to small spot-size by using an OAP. The beam is then attenuated immediately after focal-spot and relayed image then sent to a Shuck-Hartman beam-pointing sensor. The data of the diagnostics study is not presented in this thesis, however the data sets clearly showed poor pump-beam focusability and poor beam-pointing at the target-chamber. The reasons for poor focusability and poor pump-beam-pointing were explored in the fourth-generation laser-system. Experimentally found reasons for poor focusability and poor beam-pointing are presented below. All of the factors presented below were considered during the design of fifth-generation laser-system.

i. Discharge quality of LLG TWIN AMP and quality of propagating beam

The beam distortions are a critical factor resulting in bad focusability for the pump-laser beam. Poor discharge quality of the excimer-preamplifiers was one of the vital reasons for the beam-distortions. The LLG TWIN AMP excimer-amplifier has two
rectangular Polyvinylidene fluoride (PDVF) discharge tubes. Each tube has preionizer pins in two rows, parallel to the main discharge-electrode. The purpose of the pre-ionizers are to produce uniform electron-density in the KrF$^*$ gain-medium (which is a gas mixture of F$_2$, Kr, and He). Two fans, one in each tube, are mixing the gain-medium gases during the operation. The temperature of gas mixture, flow of the gas mixture, and humidity around the discharge chamber determine discharge uniformity and quality. The discharge-chambers of the LLG TWIN AMP amplifier were not in an isothermal reservoir to maintain uniform temperature inside the discharge-chamber. This caused distortions to the propagating beam. Further, the poor discharge qualities of the excimer caused erosion of the electrodes and made the electrodes surface into a non-uniform surface. The non-uniform surface of the electrodes contributed for bad discharge quality and accelerated the bad beam-profile in a very short time. During the design of fifth-generation laser-system all of these facts are carefully considered.

**ii. Shielding the beam-path**

The spatial and temporal profile-quality of a propagating laser-pulses are depending on the medium through which it propagates and stability of the medium. Further, any possible airflow across the propagating beam-line will cause beam profile distortions. The fourth-generation laser-systems had a very poor laser-beam shielding system through which pump-laser pulses were propagating before it reached the target-clusters.

The average energy at output of the KrF$^*$ amplifier (LLG TWIN AMP) was 50 mJ. Transporting 50 mJ laser pulses through air easily introduces nonlinear effects such as self-focusing, and self-phase modulation into the propagating pump-laser pulses.
Kodak photo-paper was placed at various places in the propagating beam-path, in order to record spatial-beam profile. The data showed that self-focusing effects are introduced very strong filaments to the propagating beam-profile.

A vacuum is an ideal medium for the propagation laser-pulses without introducing any distortions. It is very important to maintaining reasonable vacuum level from the final amplifier (Prometheus) output window to experimental-chamber input window in order to avoid beam distortions. In the fourth-generation laser-system, the beam-line vacuum systems were evacuated using PVC tubing system and the typical vacuum-level of the tubing system was 100 mbar. At this vacuum-level, the refractive index of the propagating medium is not unique and hence causes the self-focusing effects to introduce filaments into the beam-profile. Further, the vacuum-leaks caused strong airflow across the propagating-beam and resulted in a serious distortion to the propagating beam-profile. All of these feedbacks of the fourth-generation laser-system are considered during the design and construction of new shielding systems and vacuum systems for fifth-generation system.

iii. Pump-laser beam-propagation length

The self-focusing effect, which introduces filaments to propagating beam, is depend on the beam travel-distance and the medium through which the laser beam propagates. In the fourth-generation laser-system, laser-beam propagating distance from the Hurricane laser to the experimental-small-chamber was about 150 meters. It is obvious that the laser beam propagating such a long distance experiences very strong self-focusing effects and very poor beam pointing at target-clusters. Further, aligning the
beam for every day experiments with the same input angle for OAP was almost impossible with such a long distance. All of these facts were considered during the design of fifth-generation laser-system.

iv. Independent isolated systems

In the fourth-generation laser-system, the experimental-chamber and the seeding laser-system were built on 11 isolated, and independent, optical-tables. Therefore, a small possible misalignment in one system triggered a big misalignment inside the small-chamber. Further, the long beam-path (~ 150 m) accelerated the misalignment process inside experimental-chamber. The long beam-path provided poor beam-pointing and hence altered focal-spot quality of the pump-laser every time pump-laser interacts with target-clusters. The fifth-generation laser-system is built on a single monolith system that completely removes all of these problems.

v. Quality of optical and mechanical components

The quality of optical-component and mechanical parts also decides the beam-profile quality and beam-pointing. The surface quality of the beam-steering mirror of fourth-generation laser-system was λ/10 at 633 nm. The lenses and the windows of the LLG TWIN AMP excimer were made out of CaF₂. Polishing the CaF₂ for higher surface quality practically is a challenging project. In the fifth-generation laser-system, all the optics are carefully choose to avoid these problems.

Height of the beam-steering mirror-holder is a critical factor, which decides beam-pointing-stability. In the fourth-generation laser-system height of the beam
steering, mirror-holders were reasonable high. It is obvious that the long-arm type mirror holders will amplify any small vibration caused on the optical-tables and accelerate beam-pointing instability. A diagnostic experiment was performed to study the stability of the optical-mirror-holders. The experimental results showed that the optical-mirror-holders of the fourth-generation laser-system were performing pendulum-type motions. In the fifth-generation laser-system height of the optical-holders are reduced to a smaller length.

In the fourth-generation laser-system, all of the beam steering mirrors are attached to the conventional kinematics-mounds for the tip and tilt actions. In a typical conventional kinematics-mounds, micrometers for tip and tilt actions are perpendicular to the direction of the gravity. Therefore, the weight of the beam steering-mirror pulls down the tip and tilt plane and then misaligns the beam-steering mirrors. In the fifth-generation laser-system some of the critical conventional kinematic-mounds are replaced by specially designed beam-steering mirror-holders. In the special design, one of the micrometers for the tilt action is parallel to the direction of gravity. The micrometer will prevent the beam steering mirror tipping down due to gravity and keeping them in an extremely stable position.

vi. The ASE level inside experimental chamber

The ASE pulse produced by preamplifiers forms a low-intensity pedestal at the pulse inside experimental-chamber. This pedestal may pre-ionize or even destroy the target-clusters before the main laser-pulse arrives, resulting in null-experiment results. The beam-path in the LLG TWIN AMP amplifier is double pass off-axis cross fashion in
each tube reduces the ASE along the propagating beam as the most of ASE from discharge travels in on-axis. The ASE aperture-filter placed right after output of first amplification removed reasonable amount of ASE produced by the first amplification stage. The remaining ASE reaches the second amplification chamber and is amplified by the discharge. The final ASE level at the output of the LLG TWIN AMP amplifier was \( \sim 10 \, \text{mJ} \) while the average output energy was \( \sim 50 \, \text{mJ} \) with fresh gas-fill in the excimer. The ASE generated by the LLG TWIN AMP amplifier passed two-times through the Prometheus preamplifier in on-axis fashion and is amplified. The amplified ASE by the Prometheus reached chamber to pre-ionize the target-clusters before the main pulses reach the target-clusters. The typical ASE level inside experimental-chamber was nearly 100 mJ with the fresh gas-fill for Prometheus and LLG TWIN AMP excimer-amplifier. Prometheus is a device carefully designed to produce very low ASE. The maximum ASE produced by the Prometheus alone inside the small-chamber is less than 1 mJ for fresh gas-fill. Therefore, it is clear that preventing the ASE of excimer-preamplifier reaching Prometheus for further on-axis amplification will result to reduce the final ASE level inside experimental-chamber. In the fifth-generation laser-system, a few different approaches are being used to attenuate the final ASE level inside the experimental-chamber.

The above six-parameters are carefully considered during the design and construction of the fifth-generation laser-system. The diagram 2.3 summarizes the above parameters those are necessary to consider during the design and construction of the fifth-generation laser-system for optimizing brightness of the pump-laser.
2.3.1.1 Infrastructures Design

Based on the detail research performed on the previous laser-system, the fifth-generation laser-system is designed. In the design of fifth-generation laser-system, all the benefits of the previous laser-systems are implemented and all the drawbacks of the previous systems are either removed or modified using latest available technologies.
First of all, three big optical-tables (the sizes are 5’x10’x1’, 5’x12’x2’) and two small optical-breadboards (48”x24”x4”) coupled together and form a monolith-system. Unlike the previous system, the Hurricane seeding-laser, the entire beam steering-mirrors, beam expanding-telescope, and the experimental-chamber are on the monolith-system. Since the system is built on a single monolith-system, beam-steering mirrors and the target-cluster have same frame of reference. Therefore, unlike independent system, any possible vibration from external sources will not be amplified at the target-clusters. Even the external vibrations transferred to the monolith-system, then the vibrations might not effect the target-clusters since every single experimental-components are on a single system. Further, the height of the monolith-system is reduced from the previous 42 inches optical-table height to 36 inches in order to reduce vibration-transfer from floor to the optical-tables. Pieces of ebony woods were placed in between optical-table and laboratory floor to isolate the monolith-system from the laboratory floor vibration. The analytical diagnostic experiments performed on ebony-wood, which supported optical-tables clearly showed that the ebony plays an important role to isolate the optical-table from floor vibrations even thou we could not scientifically explain the reasons for this physical behavior of the ebony. Alignment sensitive optical-instruments such as TG FROG, Michelson interferometers, UV spectrometers are mounted on the system to test the stability of the monolith-system and the beam-pointing stability of the monolith-system. The monolith-system easily passed the test. The optical-instruments were easily aligned in the monolith-system and the aligned optical-instruments were able to keep the optical-alignment for several days without adjusting any optical-components. This confirmed the stability of the monolith-system. Figure 2.3 shows the monolith system.
As discussed previously, any possible airflow across the beam-line is one of the important factors, which damages beam-profile. Further, the airflow aggravates filamentation effects through self-focusing during the propagation of high-energy laser-pulses through air. Airtight enclosure systems are built for shielding the propagating-beam from the Hurricane output to the Prometheus input. Special gaskets were used in the enclosure design such a way that the enclosure could be filled with He gas up to atmospheric pressure. The He gas medium is suitable candidate rather than air medium to avoid any possible self-focusing effect or self-phase modulation during the propagation of high-energetic preamplified KrF* pulses. Two separate, ideal, high-vacuum systems are designed, one from first output window of the Prometheus amplification-chamber to second input window of Prometheus amplification-chamber and the second one from Prometheus output to experimental-chamber input window, to transport-high power laser-pulses without introducing any distortions. Four stainless steel vacuum-chambers and an aluminum vacuum-chamber were designed and built for keeping the beam-steering mirrors inside high-vacuum. Inside these vacuum-chambers two narrow-rails systems were built to mount the beam-steering mirror-holder. The narrow-rails reduce any possible misalignment caused by chamber deformation during the evacuation process. Stable platforms were built on optical-tables to raise the turning chamber to the beam-line height. All the beam-steering turning-mirrors after the first beam-pass of Prometheus are designed such a way to tune beam-line alignments without breaking vacuums. Vacuum stepper motors are installed to the beam-steering mirrors-holders for actuating the mirrors in order to perform beam alignment. Controls of all of these beam-steering mirrors are located close to the experimental-chamber so that experimentalist can monitor the pump-
laser-beam inside the experimental-chamber and align it by actuating each and every beam-steering turning-mirrors. There are two oil-free scroll-pumps one for the vacuum-system from Prometheus output to experimental-chamber input and the other one for the vacuum-system from Prometheus first-pass output to second-pass input are installed to make high-vacuum to transport UV high-power laser-pulses without introducing any distortions. A typical vacuum level in the tube system is ~ 25 mtorr where self-focusing and self-modulation effects are either negligible or none. The architecture of the design is presented in figure 2.3 after presented experimental-chamber and preamplifier design.

2.3.1.1.2 Experimental Chamber Design

The focal-spot quality and the perfect diagnostic alignments are the two vital parameters that decide the optimization and the characterization of the x-ray source. Easy and convenient accesses inside the chamber provide convenience during the focal-spot and diagnostic alignments. Therefore, easy and convenient accesses inside the chamber are also additional two parameters, which determine focal-spot qualities and the diagnostic alignment qualities. The previous experimental-small-chamber had access through the top of the experimental-chamber. The view ports of the small-chamber provided limited line of sight for checking alignments of focusing-setup and diagnostics. All of these facts were considered during the design of new experiential-chamber.

First of all a model of an experimental-chamber was built using cardboard for the purpose of experiencing convenient access to inside the chamber, instrumentation configuration inside chamber, optimizing size of the chamber, and reducing weight of the proposed chamber. The model proposed the following parameters
i. **Convenient access inside the chamber:** The necessary access inside the chamber for the alignment purpose should have an 24" x 36" size access door in one side of the chamber. Such a big door will make the cylindrical chamber looks like D shape and provide a convenient access for aligning diagnostics and the focal-spot monitoring-setup. The chamber will be referred from here on as the D chamber.

ii. **Size of the D chamber:** The configuration of diagnostic instrumentations and size of the optical-table of the monolith-system on which the chamber will be mounted are two factors that decide the size of the experimental-chamber. The optimized size of the D chamber, which could accommodate all the diagnostic instrumentations, and it still could be mounted on the monolith-system, is 40 inches and the high of the chamber is 24 inches.

iii. **Material of the chamber:** A lightweight chamber is an important choice to make a uniform weight-distribution on the monolith-system. The material used for the chamber is 6060 aluminum, which is an important material used to manufacture aircrafts and fuel tanks for spacecrafts. The 6060 aluminum is lightweight and hard enough to keep the high-vacuum needed for the x-ray experiments.

iv. **Type of flanges:** The chamber will have three ISO K 160 flanges (one for electrical feed-though and two for beam propagation inside chamber), one ISO CF 230 flange for Turbo pump, one ISO K 270 for gas cooling and cooling water feed-through, and one ISO K 300 for target-gas and view port. The base of the chamber will have three ISO CF 40 flanges for installing breadboard for the
diagnostics. The breadboard is completely isolated from chamber as explain in the following paragraph.

After careful research and analysis from the built sample model, the Atlas Technologies built and then delivered a D shaped aluminum chamber to UIC. Figure 2.1(b) below shows the drawing of the D chamber.

Unlike typical experimental-chambers, the D chamber has special design to isolate the breadboard from the D chamber as well as from the monolith system. The isolation breadboard designs are the following. The Atlas Technologies Inc company designed and built a half inch thickness D shaped breadboard for the D chamber. Three ISO CF 40 high-vacuum bellows (Kurt J. Lesker part number: MHT-QF-A03) attached to the bottom flanges of the D chamber. The other ends of the bellows were closed with ISO CF 40 blank flanges. Each of these flanges has a countersink at the center of the flanges. These three blank-flanges are clamped down to the monolith-system while the chamber is resting in different three legs. One end of the three threaded rods (0.75\(^*\) -16) is machined to make a sharp-point. The other ends of the threaded rods are mounted on the breadboard as shown in figure 2.1. The breadboard is mounted such a way the sharp point of the threaded rods making point-contacts with the counter-sink center-holes of the CF 40 blank-flanges. The point-contacts make a small contact on the monolith-system with the breadboard totally now isolated from the chamber. A Michelson-interferometer setup is built inside chamber to test stability of the breadboard and vibration isolation from external-vibration sources. The results from the Michelson-interferometric experiments clearly showed the breadboard is completely isolated from the chamber and partially isolated from the monolith-system. Further, any possible vibrations introduced to the
chamber or to the monolith-systems, were not transferred to the breadboard. Further, the experimental outcome presented in chapter 4 proves the success of the design and constructions. The schematic diagrams of the chamber are presented in figure 2.1 and detailed technical drawing is presented in the appendix 2.3.

![Figure 2.1: (a) Isometric view of the D chamber, (b) Breadboard setup, (c) Top view of the D chamber.](image)

**2.3.1.2.3 Preamplifier Design**

The preamplifier excimer-lasers are good candidate for reaching higher pump-power at the UV wavelength regime. The reason is the excimer-lasers are gas-lasers and they can utilize a large amplifying volume without risk of damaging the medium, which is the primary concern in crystal oscillators. Further, the excimer-amplifiers are good candidate for amplification as it is possible to use Direct Amplifications (DA) rather than CPA. However, the disadvantages for the excimers are that the typical pulse-width of excimer-lasers are nanoseconds, which is one million times longer than femtosecond pulses. Therefore, in the XRIM, excimers are being used as amplifier instead of using them as source laser in the architecture of the laser-system for the generation of
energetic-femtosecond pulses. In a typical KrF\textsuperscript{*} excimer, the time to store energy is about 2-3 ns and the entire gain lifetime is 15 ns. Therefore, it is possible to send multiple pulses throughout the amplifier volume during a single amplification cycle. In the amplification chain of the XRIM, the UV pulses are passing through KrF\textsuperscript{*} gain-medium six times, that includes four off-axis passes in excimer-preamplifier and two on-axis passes through Prometheus high-power amplifier.

A brand new KrF\textsuperscript{*} excimer-amplifier designed and built at Laser Laboratorium Göttingen GmbH (LLG), Germany. The LLG team has extensive experiences in the field of KrF\textsuperscript{*} excimer-amplifiers. The KrF\textsuperscript{*} excimer-preamplifier named as LLG 50 PRO. The previous experiences and feedbacks from the previous KrF\textsuperscript{*} excimer-preamplifiers, advantages, and drawbacks of the previous excimer-amplifiers were provided to LLG for designing purpose. LLG considered all of these facts during the design and built a new upgraded version of the LLG TWIN AMP excimer-preamplifier. The basic differences between LLG 50 PRO and LLG TWIN AMP are presented below in the next paragraph.

**Internal design to improve pre-amplified beam quality:** Both LLG TWIN AMP and the LLG 50 PRO excimer-preamplifiers in XRIM have two separate discharge-channels for four off-axis multiple-beam passes doing amplifications. In a typical KrF\textsuperscript{*} excimer-preamplifiers, in order to satisfy the necessary conditions for the preamplification, the first discharge-channel is shorter than the second discharge-channel and the distance between electrodes in the first discharge-channel is shorter than the distance between electrodes in the second discharge-channel. The distance between electrodes decides the beam-size, and hence the amplification. The electrodes distance in the LLG 50 PRO is
slightly larger than the LLG TWIN AMP, which makes the beam size at the output of the amplifier slightly larger than the previous LLG TWIN AMP excimer. The beam size at the outputs of first and second channels of LLG 50 PRO is presented in the picture in figure 2.2.

In KrF* excimers preionization mechanism plays crucial the role in producing a uniform glow-discharge and in avoiding arcs in the discharge. The preionization mechanism is the process, which seeds the discharge volume uniformly with electrons prior to the initiation of the main discharge. Electron densities of $10^7$ to $10^9$ cm$^{-3}$ are required to produce uniform arc free discharge. In a typical KrF* excimer-preamplifiers, similar to the ones in XRIM, preionization pins are arranged in a row adjacent to the discharge electrodes, and the preionization mechanism happens 10 ns earlier than the main discharge. The electrode systems commonly used for the excitation of high-repetition rate excimer-lasers falls into two categories: Solid electrodes with side preionization spark-arrays or corona bars and combinations of solid and screen electrodes with preionization sources located behind the screen. The first type of electrode system is used in the excimer-preamplifiers such as LLG TWIN AMP and LLG 50 PRO of XRIM and the second type of electrode systems are used in the Prometheus amplifier. In the design of the LLG 50 PRO, numbers of preionization pins are higher than number of preionization pins in the LLG TWIN AMP. In addition to an increased number of preionization pins, a set of pre-pre-ionization pins are added to system to increase additional uniformity to the preionization process. The parameters that affect the preionization (such as threshold value, seeded electron density and uniformity effects) are heavily dependent on the design of the discharge system such as type of electrode and its
profile, gas pressure, duration of preionization, electron loss process, time delay between
preionization and main discharge, rise time of the electron voltage as well as on overall
discharge system geometry [39]. All of these parameters are carefully considered during
the designs of the LLG 50 PRO in order to optimize the discharge quality hence the
beam-profile quality.

In order to improve the discharge qualities further and in order to remove all the
filaments formed during the main discharge and pre-ionizer discharge, a new idea of
auxiliary electrodes introduced to the discharge reservoirs. LLG 50 PRO consists a pair
of auxiliary electrodes installed to the side wall of the discharge-reservoirs such a way
that the electrodes faces each other and clear the off-axis beam passes and perpendicular
to the main discharge electrode and pre-ionizer pins. Further, the designs enable a change
in the distance between auxiliary electrodes by rotating alignment screws located outside
of the reservoirs. A complete rotation of the screws move the electrodes back to original
positions. It was experimentally observed that manipulating the auxiliary electrode
distances improve the quality of homogeneity of the amplified beam while it decreases
the output-energy of the excimer. Therefore, in LLG 50 PRO all four auxiliary electrodes
are tuned to produce highest homogeneity pre-amplified beam with reasonable output
energy, which is enough to produce optimized output energy from Prometheus.

The positive pressures level of each excimer reservoirs and the pressure
difference between both reservoirs determine the time delay between first stage of
amplification and second stage of amplification. The percentage of He, F, and Kr gas-
mixture in the first and second reservoirs further determines the discharge quality and the
final output energy of the excimer-preamplifier. In the LLG 50 PRO excimer-
preamplifier the optimized operating pressures of the first reservoir (short tube) is 2.4 bars with 5 % F and He mixture (4.71 % ≤ F ≤ 6 % and rest is He), 6.25 % Kr, and 88.75 % He and the optimized operating pressure of the second reservoir is 1.6 bar with 7.5 % F and He mixture (4.71 % ≤ F ≤ 6 % and rest is He), 9.375 % Kr, and 83.125 % He.

All four windows of the LLG 50 PRO are carefully selected to reduce beam-distortion and optical-chirp effect when the 248.6 nm pulses pass through the windows. Since the MgF$_2$ has a higher order surface quality compared to the CaF$_2$ windows, all four windows of the LLG 50 PRO windows are MgF$_2$. Further, the thicknesses of the windows are slightly reduce to 5 mm from previous thickness of 10 mm in order to reduce optical-chirp effect on the UV pulses when they passes through windows.

The LLG 50 PRO was tested at LLG before deliver to UIC. The seeding-pulses used for the test at LLG were similar to seeding UV pulses generated in XRIM from the Hurricane and tripling crystals. During the test at LLG a Transient Grating (TG) FROG was used at LLG to measure pulse-width at the output of the excimer. The pulse-width was ~ 350 fs. The peak power was 0.14 TW at 40 mJ energy and brightness of the excimer-preamplifier was in the order of ~ 10$^{20}$ Wcm$^{-2}$ sr$^{-1}$). The dimension of the output beam was 39 mm vertically and 28 mm horizontally.

**External design to improve pre-amplifier beam quality:** As discussed above discharge quality of the main electrode during the excimer-amplification process determines the beam-profile quality. Maintaining uniform reservoir-gas temperature of the excimer and removing the humidity around the excimer-reservoirs and capacitors banks improves drastically discharge quality of the main electrodes, and hence the beam-profile quality.
The previous excimer-preamplifier did not have any temperature or humidity controls around the gas-reservoirs and the capacitors banks. Two closed-loop water-circulating systems are designed and built to maintain the temperature of the gas-reservoirs and the Thyratron switch, which controls the discharge mechanism. The closed-loop water-circulation systems has a heat-exchanger, and a water circulating system to maintain constant-temperature around the reservoirs. The diagrams of the closed loop water circulating systems are presented in the appendix 2.4. A commercial dehumidifier was installed using a duct system to remove the moisture air around the reservoir and capacitor banks of the LLG 50 PRO excimer-preamplifier. The dehumidifier runs the moisture-free air through the excimer-amplifier enclosure to replace moisture air around the reservoirs and the capacitors banks of the LLG 50 PRO excimer.

The internal and external designs resulted in a decrease of the ASE level by factor of 10 compare with the LLG TWIN AMP. The ASE level of LLG 50 PRO is 1 mJ for a fresh-gas fill with 100 mJ output energy while it was 10 mJ for LLG TWIN AMP with fresh gas fill and output energy of 50 mJ. Figure 2.2 below compares the beam profile quality of the LLG 50 PRO and the LLG TWIN AMP and confirms that the improvement of beam-profile quality.
Selection of optics to improve pump-beam quality: A new optical-architecture is built on the monolith-system for sending seeding UV pulses to the pre-amplification of LLG 50 PRO and then for Prometheus pre-amplification. The Hurricane Ti: Sapphire ultrafast laser-systems, tripler optical-setup, and beam-expander optical-setup of the fourth-generation is upgraded and recycled into the fifth-generation laser-system. Optical qualities of all the optical-components are improved in the new system. All the optical-components are procured from LASEROPTIK GmbH. All of beam-steering mirrors are dielectric coated for 248.6 nm at 45 degrees with surface qualities of $\lambda/20 @ 633$ nm. All the windows of the spatial filters and LLG 50 PRO are made out of MgF$_2$ with $\lambda/10 @ 633$ nm surface qualities. The reason for the selection of MgF$_2$ rather than CaF$_2$ is that the CaF$_2$, compared to MgF$_2$, has an isotropic structure, lower hardness. Further, CaF$_2$ is sensitive to thermal shocks and achievable possible higher surface-smoothness is $\lambda/4 @ 633$ nm. One of the drawbacks of MgF$_2$ compare to the CaF$_2$ is that MgF$_2$ is birefringent. In order to minimize the birefringence effect, the MgF$_2$ windows are fabricated with the optic-axis as a symmetry axis (c-cut). Therefore, during the installations of the MgF$_2$ optic, require the help of a UV polarizer, the optical-axis of the transmitting optics are
carefully aligned to minimize the birefringence effect. All the lenses are positive Plano-concave and details of their focal lengths and the material are presented in the next paragraph. The gratings of the pulse compressor are Littrow grating with 2100 lines per mm and 74% efficiency at 250 nm.

**Seeding UV beam-line for pre-amplifications:** The beam-line path for the first four preamplification stages of the LLG PRO 50 is presented in figure 2.4. The beam-line is built with relay-imaging principles [40]. The relay imagining is restores the input beam-profile on the output of the LLG 50 PRO regardless whatever happens to the beam profile in-between the input and output. The focal-length of all the Plano-concave lenses and all other optics are carefully chosen to satisfy the relay-imaging criteria. In the relay-imaging setup of XRIM, the beam-profile of the Hurricane laser output is projected to the output of the second-stage amplification of LLG 50 PRO and then the output beam-profile of the second stage amplification is projected to the final output of the LLG 50 PRO. The details of beam-path lines are presented in figure 2.4.

As presented in the fourth-generation laser-system the 745.8 nm wavelength Hurricane laser pulses are tripled up to 248.6 nm using tripling techniques. The 14 cm beam height of the tripled up UV beam from tripling crystal is lowered to 10 cm using a periscope setup. The polarization of the beam at the input and output of the periscope is in the vertical direction, relative to the optical-table. The beam is then sent to an MgF₂ Plano-concave lens, of 496 mm focal-length, to focus the beam inside vacuum and thereby create a point source. The point source removes any possible distortion in the beam and further increases homogeneity of the beam. The emerged beam from the
vacuum is collimated using an MgF$_2$ Plano-concave lens of 1731 mm focal-length and then the beam is sent to the LLG 50 PRO for the first of two amplifications, in an off-axis fashion. It has 2 degrees input angle for first pass, and 3 degrees input angle for the second pass. The energy at the output of second-amplification stage is about 14-17 mJ average energy and 25mm x 20 mm beam size. During the beam passes through the gain-medium, and the windows the -pulses experience to the spectral and temporal broadening and pulse-chirping. In order to compensate the spectral and temporal broadening the amplified beam is then sent to a grating-pair setup. The output from the grating-pair is passes through a fused silica Plano-concave lens of 2721 mm focal-length to focus the beam to a vacuum-aperture of 270 μm diameter for removing the ASE from the first two-amplifications of the LLG 50 PRO excimer-preamplifier. The lens (the one with 2721 mm focal length) is mounted on a special rotating-stage in order to produce a diffraction-limited focal-spot at the vacuum-aperture. A special focusing-device, FSMOS, together with adjustment techniques (presented in the chapter 2.3.1.2.2) used during the installation of the lens to produce diffraction-limited focused-spot at the aperture. The diverging beam from the aperture is slowly raised to 11 cm beam-height using two beam-steering mirrors, and then collimated using a fused-silica Plano-concave lens, of 2953 mm focal length. The beam is then sent for the second two-passes of the LLG 50 PRO amplification, at an off-axis fashion with 2 degrees for first-pass and 3 degrees for the final-pass. The beam size at the final stage of amplification is 45 x 35 mm and average-energy of 70 mJ for fresh gas fill.
The amplified beam from LLG 50 PRO is then sent to the BET, which consists of two spherical UV mirrors for the expanding the 39 x 28 mm rectangular beam to 9 cm x 10 cm size rectangular beam. The expanded-beam is then sent to Prometheus for the first amplification. The Prometheus first-amplified beam is then sent to the second-discharge chamber of the Prometheus for the final amplification, by two 6 inch x 5 inch size dielectric beam-steering mirrors, termed in this thesis, as Big Mirror 1 (BM1) and Big Mirror 2 (BM2). The final amplified-pulses then sent to the experimental-chamber with three beam-steering dielectric-mirrors, termed in this thesis as, Big Mirror 3 (BM3), Big Mirror 4 (BM4) and Big Mirror 5 (BM5). The beam-line path is presented in the figure 2.4. The beam-path alignment procedures are presented below.

2.3.1.1.2.4 Pump-Laser Beam Alignment Procedure

The pump-laser beam alignment procedures are divided into two separate-regions one from output of tripling crystal to input of Prometheus, the other one is from Prometheus input to the OAP.

During the initial installation of the fifth-generation laser-system, all the optics from tripling box to the Prometheus input are carefully aligned to produce aberration-free perfect beam-line path from the Hurricane to the BET. There are seven irises installed in the beam-line path to define the beam-line path. An iris is installed before the first periscope to define the output-beam from tripling crystals, and there are irises installed before each and every lens to define beam-line. Further, there are irises installed to define input and output beam-line path for the vacuum-aperture unit, and finally an iris is installed to define the final output beam of the LLG 50 PRO.
The beam-line path from output of first discharge-chamber of the Prometheus to OAP is designed such a way the beam alignment could be performed inside a vacuum. There are two different guided lasers installed in the system to align the pump-laser beam and the OAP setup to reproduce the beam-alignment path exactly same for the day-to-day experiments. The first guide laser is mode-locked diode laser-pointer (wicked lasers: S3 Artic series) installed into the system such a way that the laser-beam follows the pump-laser beam collinearly from the BM1. The second guide laser is an Argon-ion (Ar⁺) laser (Coherent, Inova 300) installed into the system such a way it follows the pump-laser beam collinearly from the BM3. The guided laser beams paths are shown in figure 2.4.

A special designed iris is installed immediately after the BM2. The iris designed enables the experimentalist to introduce the iris into the propagating beam-line path without releasing vacuum of the beam-lines tube. Once the alignment is ready the iris could be lift it up without breaking the vacuum. At the position where the iris is introduce into the beam-line path, the center of the iris is matched with the center of the pump-laser beam if the beam-line is in alignment before the iris. There are two irises installed inside the D chamber to definite the input beam-line path to OAP. If the input beam to the OAP is aligned, the centers of both irises will match with center of the laser beam. The alignment procedure of the whole pump-laser beam paths are presented below.

I. The first step is to align the beam from Hurricane laser to the output of the BET. During the daily beam-alignment, the beam positions on all the irises are carefully checked from tripling output to the BET output. The first iris will be closed to generate diffraction fringe-patterns. In a perfect alignment, the diffraction pattern will be a collinear to the center of all irises. If the beam is not collinear to the any irises
the beam-steering mirrors immediately before that particular iris will be tuned to make the diffraction pattern collinear to that iris. The same procedures will be repeated for all the irises up to the output of BET. The iris immediately after the BM2 will be dropped and co-linearity of the beam on the iris will be checked inside D chamber (shown in figure 2.4). If it is not collinear, the alignment procedures will be repeated again until co-linearity is obtained on the iris, which is immediately after the BM2. The co-linearity of the iris and the pump-laser beam will guarantee the input beam-line path to the first discharge-chamber of the Prometheus is aligned.

II. The second step is to align the beam from output of BET to the OAP.

II.I. The beam-line path of the second guided-laser is checked with the two irises inside the experimental D chamber. If the beam is not collinear to both irises the BM 4 and BM 5 will be actuated inside vacuum until the guided beam is collinear to both irises.

II.II. During the installation of the fifth-generation laser-system, a perfect beam-line path was aligned from output of BET to OAP. Using both guided lasers, beam-points were built, as explained below, to define perfect beam alignment positions of the BM1, BM2 and BM3. A reflection of the first guided-laser from the back side surface of the BM1 is sent to crosshairs, which defines the perfect alignment of the mirror. The reflected first guided-laser beam from BM2 is sent through the BM3 to crosshairs, which defines the perfect alignment of the BM2. Finally, reflection of the second guided laser from the backside surface of the BM3 is sent to crosshairs, which also defines the perfect alignment of the BM3. Once the BM4 and BM5 are aligned perfectly, as explained in the II.I, the beam-pointing of the
BM1, BM2, and BM3 are checked at respective crosshairs. If the beam is not aligned on the crosshair the appropriate mirrors will be moved to make perfect alignment.

II.III. Pump-laser beam-line path will be checked on both irises located inside D chamber. If it is not aligned to both irises, move the BM2 and BM3 to make the pump-laser beam collinear to both irises. Now all three laser beams the first guide-laser, second guide-laser beams and the pump-laser beam should be collinear (shown in figure 2.4). The co-linearity of the three laser beams confirms the correct input beam-path for the OAP.

The beam-line path alignment procedures, presented above, enable the experimentalist to reproduce the OAP input beam-line path direction to the order of micron accuracy on day-to-day OAP alignments. Table 2.1, below, compare all the laser-systems of XRIM and shows that the 5th generation laser-system is providing optimized pump-power in controllable fashion.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>1st generation</th>
<th>2nd generation</th>
<th>3rd generation</th>
<th>4th generation</th>
<th>5th generation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average Energy (mJ)</td>
<td>5</td>
<td>220</td>
<td>300</td>
<td>350</td>
<td>350</td>
</tr>
<tr>
<td>Pulse-width</td>
<td>10 ps</td>
<td>600 fs</td>
<td>~ 500 fs</td>
<td>~ 400 fs</td>
<td>~ 350 fs</td>
</tr>
<tr>
<td>Wavelength (nm)</td>
<td>193</td>
<td>248.6</td>
<td>248.6</td>
<td>248.6</td>
<td>248.6</td>
</tr>
<tr>
<td>Power (W/cm²)</td>
<td>(10^{13})</td>
<td>(10^{16})</td>
<td>(10^{19})</td>
<td>(10^{20})</td>
<td>(10^{21})</td>
</tr>
<tr>
<td>Size of the laser-system</td>
<td>N/A</td>
<td>7 optical-table</td>
<td>7 optical-table</td>
<td>7 optical-table</td>
<td>3 optical-table</td>
</tr>
<tr>
<td>Distance from the oscillator to target chamber (m)</td>
<td>Not available</td>
<td>200</td>
<td>200</td>
<td>150</td>
<td>10</td>
</tr>
</tbody>
</table>

Table 2.1: Comparison of all laser systems of XRIM [34], [35], [36], [37], and [38]
Figure 2.3: Architecture of the XRIM laboratory. The figure shows all-important optical-components are built on monolith-system.
The following section 2.3.1.2 discusses alignment of the OAP.
2.3.1.2 Focusing Devices

Producing diffraction-limited focused-spot on target-clusters is necessary conditions to achieve enough pump-power for the generation of the ultrabright x-ray source. In the x-ray XRIM a carefully aligned OAP mirror focus the pump-laser beam to produce diffraction-limited focal-spot on the target-clusters. In order to align the OAP to produce diffraction-limited focal-spot, OAP should be moved in five independent axes (y, z, rotation, tip, and tilt) with respect to the input-beam and the focused-spot should be monitored, during the alignment process, using an aberration-free imaging setup. In the following section, design of five-axes OAP positioner, design of focus-monitoring optical-systems, alignment procedures are presented.

2.3.1.2.1 Design of Off-axis Parabolic Positioner

The Arabian physicist, Ibn Sahl, first described parabolic mirrors in the 10th century. It was later described again by Ibn al-Haytham (Alhazen) in his famous book of optics in 1021. Sir Isaac Newton made a parabolic reflecting telescope in 1689 by replacing the previously used spherical mirrors in the telescopes. John Hadley introduced parabolic mirrors into practical astronomy in 1721 when he used one to build a reflecting telescope with very little spherical aberration. Even before the parabolic mirrors found its application in the field of telescope, lighthouses commonly used parabolic mirrors to collimate a point of light from a lantern into a beam, before being replaced by more efficient Fresnel lenses in the 19th century.

Precision Asphere Inc. manufactured the off-axis parabolic mirrors of the XRIM laboratory. As the first step of the OAP construction, large enough "parent" or "mother" on-axis paraboloid substrate was constructed from zerodur and polished to the \( \lambda/10 @ \)
633 nm surface quality. The OAP’s are machined and removed from the parent in a specific distance from the optical-axis of the parent. The removed OAP’S are coated with dielectric-material for optimizing reflections of the incident 248.6 nm laser beams. The detail specifications of the OAP’s are presented below.

- Substrate material is zerodur.
- Distance between optical-axis of OAP and the optical-axis of parent paraboloid is 3 inches (7.62 cm).
- Off-axis angle is 30 ± 0.06 degrees.
- Focal length f is 20.32 ± 0.02 cm.
- Diameter of the OAP d is 10.66 cm.
- Clear aperture of the OAP is 10.16 cm
- Surface Roughness: < 2 nm RMS
- F number at effective beam-size of 9.5 cm (f/d) is ~ 2

A line, which is going through the center of the OAP and parallel to the optical-axis of the parent paraboloid is defined, in this thesis, as the optical-axis of the OAP. The OAP, like other imaging optics, produces third order aberration such as spherical aberration, coma, and astigmatism for any possible misalignments. In focusing-systems, these aberrations manifest as changes in the focused-spot energy distribution. In general, diffraction-limited alignment is defined as the alignment, which minims the third order aberrations to one quarter of the incident wavelength (λ=248.6 nm/4). In the diffraction-limited alignment, the optical-axis of the OAP coincidence with the axis of the incident beam and the plane of focal-line and the OAP optical-axis, coincide with the horizontal central plane of the incident beam. Therefore, in order to make the axes and planes...
coincide the OAP positioner should have to have five independent axes (y, z, tip, tilt, and rotation) of freedom.

Five independent-axes OAP positioner is designed and built using Newport corporation products a kinematic tip and tile stage (part number M-37S), a x, y, z linear-stage (part number 461-XYZ-M), and a rotation stage (part number M-481-A-S). All the stages and actuators are carefully chosen to control the alignments inside a vacuum during the experiments. Vacuum compatible actuators (Newport Corporation, part number: NSA12V6) are installed on the stages to control the linear, angular and rotation motions inside the vacuum. The minimum linear movements of the actuators are in the order of nanometers, the minimum angular rotation of the tip, tilt stages are in the order of micro-radians, and the minimum rotation of the rotation stage is in the order of micro-radians. The backlashes of actuators are nearly zero and reproducibility of the actuator and the stages are one hundred percent. Figure 2.5 below shows design of the five-axes positioner along with the OAP.

The following section is discussing a device, which monitor the focal-spot quality produced by the five-axes OAP positioner.

2.3.1.2.2 Design of Optical Setup for Monitoring Focal Spot Quality

An aberration-free optical-setup is built to monitor focal-spot quality produced by the five-axes OAP positioner. The optical-setup is termed, in this thesis as Focal-Spot Monitoring Optical-Setup (FSMOS). The FSMOS consists of a UV micro-spot focusing-objective mounted on a five-axes positioner and a CCD camera mounted on a five-axes positioner. The micro-spot focusing-objective is a Thorlabs corporation product (part
number LMU-15X-248) and the details specifications include 20 times magnification, 4 mm working distance, and 0.4 Numerical Aperture (NA). The CCD camera is a product of Lumenera Corporation (part number LU 125). The CCD camera pixel size is 5x5 μm, and the chip is coated with phosphor for optimizing the sensitivity to the 248.6 nm wavelength.

A Newport corporation 3-D stage 9062-XYZ-PPN and Thorlabs corporation objective mount KM100R are combined to built a five-axes positioner for micro-spot focusing-objective. The micro-spot focusing-objective mounted on the five-axes positioner will relay image magnified aberration-free image of the focused-spot on the CCD camera chip. The CCD camera five-axes positioner is built using a Newport corporation 3-D stage 460A-XYZ and a Thorlabs corporation kinematic platform-mount KM200B. The five-axes positioner is keeping the CCD camera-chip parallel to image-planes of the magnified image. The optical-setup of the focused-spot monitoring is presented below in figure 2.5.

The OAP is focusing the pump-laser beam to the target-clusters and all the diagnostics are aligned around the pump-laser focused-spot to visualize the physical process during the laser-matter interactions. During the focal-spot quality check, the micro-spot focusing-objective has to be mounted along the pump-laser beam-line in the forward on-axial direction (x-axis in figure 2.5) and the front lens of micro-spot focusing-objective should be few millimeters away from the focused-spot. Therefore, all the experimental diagnostics need to be removed to fit the FSMOS. Removing all the diagnostics setups and realigning them not only time consuming procedures but also produces different diagnostics alignments for day-to-day experiments. Exactly same day-
to-day diagnostic alignments guarantee the reproducible datasets and enable the experimentalist to compare the datasets. An optical-setup named “equivalent focus-setup” was built to avoid removing all the diagnostic during the OAP alignment for producing diffraction-limited focal-spot. The equivalent focus-setup design consists of linear-pinion stage and a zero degree UV reflective mirror. As shown in figure 2.5 the equivalent focus-setup is installed, between OAP and the focused-spot spatial-location, where it does not interfere with any other diagnostics. In the design of equivalent focus-setup, the mirror mounted on the linear-pinion stage, at its raised position, diverts the apex of the focusing beam cone to a location where the focus-monitoring setup finds enough room to fit inside D chamber without interfering with any other instruments. Once the OAP is aligned to produce diffraction-limited focal-spot, the linear-pinion stage will be lowered so that the diffraction-limited focused-spot will fall on target-clusters for laser-matter interactions. Figure 2.5 shows the equivalent focus-setup along with the beam-line path. In future, the design will be upgraded to align the OAP without breaking vacuum of the D chamber to produce diffraction-limited focused-spot.
2.3.1.2.3 Alignment Procedure for OAP Alignment

The OAP alignment for producing a diffraction-limited focused-spot has two separate alignment procedures. The first alignment procedure aligns the optical-axis of the OAP to the incident-beam axis. The second alignment procedure aligns the plane, which has the optical-axis of the OAP and the focal-line, with the horizontal plane of the incident incoming pump-laser beam. Table 2.2 below shows in detail the actions of the actuators of the OAP positioner. Shown in the above figure 2.5 and figure 2.12 there are two virtual-Cartesian coordinate system that is defined inside the D chamber, for the purpose of data-analyzing and diagnostic alignments. The origin of the first Cartesian coordinates is placed 2 mm above center of nozzle. The x-axis is along the pump-laser beam with positive direction is direction of pump-laser beam. The y-axis lies in the horizontal plane and the z-axis lies in the vertical-plane. The right-hand rule defines the
direction of the y-axis and z-axis as shown in the figure 2.5. Further, a red laser-diode is placed inside the chamber D as shown in the figure 2.12 for diagnostic alignments. The laser-diode beam is perpendicular to the focused pump-laser beam, passing through a point where OAP focuses the pump-laser. In other words, the red laser-diode beam is the y-axis of the virtual Cartesian coordinate system. The red laser-diode is a product of Edmund optics (part number: NT83-822) and the wavelength of the laser is 635 nm. The other virtual-Cartesian coordinate system is shown in the figure 2.5 and it describes the incoming pump-laser beam to OAP and OAP positioner.

<table>
<thead>
<tr>
<th>Actuator No</th>
<th>Actuating direction</th>
</tr>
</thead>
<tbody>
<tr>
<td>Actuator 1</td>
<td>$X_B$: Beam axis of incoming pump-laser beam</td>
</tr>
<tr>
<td>Actuator 2</td>
<td>$Y_B$: Perpendicular to the direction of incoming pump-laser beam and actuate in the horizontal $X_B, Y_B$ plane.</td>
</tr>
<tr>
<td>Actuator 3</td>
<td>$Z_B$: Perpendicular to the horizontal $X_B, Y_B$ plane.</td>
</tr>
<tr>
<td>Actuator 4</td>
<td>Tip: Tipping about the $Y_B$ axis.</td>
</tr>
<tr>
<td>Actuator 5</td>
<td>Tilting: Tilting about the $Z_B$ axis.</td>
</tr>
<tr>
<td>Actuator 6</td>
<td>Rotation: Rotating the OAP about optical-axis of the OAP.</td>
</tr>
</tbody>
</table>

Table 2.2: Actuator labels and direction of motions

The step by step procedures for OAP initial alignment is presented below

I. Install OAP custom designed mask on the OAP such a way the optical-axis of the OAP is going through the center of mask. The mask has crosshairs on its front at the center and there is a flat aluminum mirror in front of the mask.

II. After a beam-line path alignment, actuate the OAP by using the motorized actuators 1, 2, and 3, to align the center of the beam to the crosshairs of OAP mask.
III. Actuate the actuators 4, and 5 and coincide the incident-beam and the reflected beam from the mirror mounted on the OAP mask. These two procedures nearly align the axis of the OAP to the axis of the incident-beam.

IV. Remove the OAP mask and rotate the OAP using the actuator 6 until the incident beam height and the OAP reflecting beam height are same. This procedure makes sure the plane, which has the optical-axis of the OAP and the focal-line, coincide with the horizontal-plane of the incident-beam.

The final OAP alignment procedure for producing a diffraction-limited focal-spot are briefly presented below.

I. Raise the linear-pinion stage and divert the beam to the monitor the focused-spot using the FSMOS.

II. Tune the Hurricane laser to run at 5 Hz repetition-rate and make sure the distance between front end of the micro-spot focusing-objective and the focused-spot distance is more than 4 mm. Start the CCD camera software and watch the magnified image of the focused-spot in the computer monitor. Use the appropriate Neutral Density (ND) filter placed between CCD camera and the micro-spot focusing-objective. Move micro-spot focusing-objective and make sharp image.

III. Actuate actuators 1 to 5 (not 6) and to get the similar image shown below in the diagram 2.4.1.

IV. Actuate actuator 4 and rotate the image shown in diagram 2.4.2.

V. Actuate actuator 5 and make a circular shape focused-pot shown in diagram 2.4.3.

VI. If there are strong wings observed in the focused-spot, check the spatial beam-profile at the output of the LLG 50 PRO preamplifier. The fringe patterns, caused
by the LLG 50 PRO electrodes, are producing the wings. Remove the fringes and repeat the procedure from I to V

VII. Remove micro-spot focusing-objective. Start the guide laser 2 and block the pump-laser. Mark a point where the laser beam hits after passing through micro-spot focusing-objective holder.

VIII. Install back the micro-spot focusing-objective. If the beam does not match with the above-mentioned mark, move the objective to match with the mark.

IX. Install an aperture in the focal-plane to beam passes through the aperture. Carefully look on the aperture holder for back reflected beam from the lenses of the micro-spot focusing-objective. If the back reflection-spots are not matching with the incoming beam, use tip and tilt kinematic actuators of the objective to match the reflective-beam with the incoming-beam.

X. Go back to setup I and repeat the steps from I to IX until a diffraction-limited focal-spot is reached.

![Diagram 2.4: Focused-spots variations during the OAP alignment procedure.](image)

Optimizing brightness of the realized ultrabright x-ray sources is one of research goals of this thesis. Optimizing the brightness of the pump-laser along with stable beam pointing on the target-cluster are two of the fundamental condition to optimize the x-ray source. The presented focused-spot details such as size, intensity irradiance profile in the
section 3.2.1 of chapter 3 clearly shows the success of the design. The comparison of the fifth-generation laser-system, presented in the table 2.1, with previous laser-systems further confirms success of the design of the laser-system and its related instruments. The beam pointing instability produced by the laser-systems and instruments are experimentally measured inside the D chamber by recording consecutive spatial position of the focused-spot on target-clusters. The experimentally measured results of the beam-pointing instability of focused-spot, presented in the picture 2.6, clearly shows that the beam pointing instability of the focused-spot is less than the focused-spot size that was the goal of the design.

2.3.2 Apparatus Design to Optimize Target Clusters

In XRIM, the target-clusters with the density closer to the density of the solid-target is produced by pulsed injection of target-gas from a high-pressure reservoir through a sonic-nozzle orifice into the vacuum D chamber. The cluster formation mechanism, and estimation of the clusters-size and density of the Xe and Kr clusters are presented, briefly, in section 3.3 of chapter 3.
Gas-puff-targets have a number of substantial differences and advantages when compared to solid-targets. A fundamental difference between gas-puff and solid targets is that the gas-puff provides the gain-medium at a density closer to the lasing conditions [17]. Further, the plasma produced from a gas-puff-target differs substantially from the plasma produced from a solid-target. The strong density gradients, typical for plasmas from a solid-target, do not exist in the plasmas from a gas-puff, resulting in a flat electron density profile [20]. Therefore, the gas-puff provides better control over the density and minimizes gradients in the plasma. Additionally gas-puff-target, instead of a solid-target, offers the advantage of developing a high-repetition rate ultrabright x-ray source with no target debris production.

The very first nozzle produced target-clusters in the D chamber was a commercial Parker Hannifin corporation 1.5 mm orifice diameter circular nozzle. The nozzle then replaced with a custom designed sonic-nozzle. The details of the custom designed nozzle and characteristics of the nozzle are presented in reference 41. The sonic-nozzle, which produced target-clusters for the experiments presented in thesis, is a hybrid sonic-nozzle termed in this thesis as Double Gas Sonic-Nozzle (DGSN). The reasons for the replacement of the previously designed sonic-nozzles by the DGSN are already presented in the section 2.3.

The DGSN is a hybrid redesigned double gas sonic-nozzle from the commercial product of Spraying Systems Corporation (part number: AA10000JJAU-VI). An additional custom designed sonic-nozzle was introduced to commercial nozzle along with other modifications. In the DGSN configuration, the nozzle sprays target-gas at the center while it sprays the low-Z gas (such as He, and N) at the annular region of the target-gas.
The low-Z gas compress the primary target-gas into a small volume with the appropriate backing pressures of both gases and appropriate pulsing time of both gases. Figure 2.7 below shows the schematic diagram of the DGSN. Appendix 2.5 presents schematic timing diagram.

Figure 2.7: Schematic diagram of the DGSN and the sonic-nozzle alignment target. All dimensions are in mm. Scales are in mm.

On the purpose of characterizing the DGSN, the designed DGSN introduced to the one arm of a pre-built Mach-Zehnder interferometer. The Mach-Zehnder interferometer design and theory for characterizing gas-flow pattern of sonic-nozzles are presented in reference 41. In the characterization experiments the backing pressure of the primary target gas (Xe) was kept in the typical experimental backing pressure 150 psi and the backing pressure of secondary low-Z gas (N) systematically varied until the interferogram shows the primary Xe target-gas is trapped inside a smaller volume with definite barrier between primary and secondary-gases. A sample experimental interferogram results from the Mach-Zehnder interferometer setup for the DGSN characterizations are presented below in figure 2.8.
During the experiment presented above, the backing pressures of both gases were kept constant and only the delay time between both gases changed. The interferogram of the DGSN, presented in the above figure 2.8, clearly shows the success of the DGSN design.

The DGSN was installed in a three-axes positioner (Newport Corporation, Part number: 562-XYZ-LH). Three vacuum compatible actuators (Newport corporation, part number NSA 12 V6) move the sonic-nozzle in 0.5 micrometers steps the experiment for aligning the target-cluster to the appropriate position of the pump-laser focused-spot in order to optimize brightness of the ultrabright x-ray source. In a typical experiment, an x-ray aperture camera views the target-area with appropriate filter, which blinds all the radiation other than x-rays. The x-ray fluoresce images of the channels are recorded with an aperture x-ray camera presented in figure 2.9. This confirms that the Xe molecules are trapped inside a smaller volume and the enhancement of the x-ray yield compare to the single-gas sonic-nozzles. The details result of x-ray yield are presented in the section 4.4 of chapter 4. In the experiments, the secondary gas was pulsed 4 ms after the primary Xe gas was pulsed.
Previous experiments performed in the small-chamber proved that the cooling the Xe target-gas results in an enhancement of the x-ray signals. A liquid nitrogen cooling system is to cool down primary and secondary-gas system to the same lower temperatures. All the o-rings of the DGSN are replaced with silicone o-rings for better sealing at low temperatures. The cooling designs consists of copper tubes, which wrapped to the DGSN and run through a liquid nitrogen cold bath to cool down both the primary and secondary low-Z gases to same low temperatures. The lowest temperature, which the DGSN operates without any leaks is 250 degrees Kelvin (K). The results of temperature versus x-ray signal strength (or x-ray yield), presented in reference 42 clearly show the
enhancement of the x-ray signal at low temperatures and also the success of the cooling system design.

2.3.2.1 Nozzle Alignments Procedure

A nozzle alignment target, shown in the figure 2.7, is designed to align the nozzle. The nozzle alignment target is a key tool for not only an alignment of the DGSN but also for alignment of all the diagnostic. The target is mounted on nozzle orifice and the focused preamplifier LLG 50 PRO beam will illuminate the target. After 4 or 5 shots the target is removed and then inspected. If the laser burn mark is not at the crosshairs of the target, the DGSN is then moved using the nozzle positioner guided by the observation. The above procedure is repeated until the laser focus hits the crosshairs. After this alignment, the center of the target gas nozzle orifice is in the x-axis (x-axis is described in figure 2.12).

The goal of aligning the nozzle orifice center to the focused-spot spatial location (origin of the Cartesian coordinate in the figure 2.12) is realized by splitting the incoming UV beam into three beam by introduction of a plate that has three holes. The preamplified beam is then fired at the nozzle target. After several shots the target is removed, and then inspected with a microscope. If there are three burned marks on the target then repeat the procedures until a single burn mark is achieved. At the end of this procedure nozzle orifice is aligned to focused-spot.
2.3.3 Design of Experimental-Diagnostics for Optimizing X-ray Source

In experiments, diagnostics act as eyes of experimentalist to visualize the physical process-taking place during the experiments. In experiments to generate and characterize an ultrabright x-ray source at XRIM, there are eight diagnostics; (1) triplet-aperture x-ray camera, (2) axial-von Hámos spectrometer, (3) transverse-von Hámos spectrometer, (4) Thomson scattering, (5) pump-laser energy, (6) target-gas backing pressure, (7) secondary-gas backing pressure, and (8) target-gas temperature. These 8 diagnostics monitor the target-area and record single-shot data every time the pump-laser interacts with the target-clusters. The diagnostics instruments of the experiments presented in this thesis are constructed using the latest technologies available for diagnostic design and construction. In the following sections, each and every diagnostic design and alignment procedures are presented briefly.

2.3.3.1 Design of Triplet Aperture X-ray Camera

The direct imaging of x-ray source is a challenging since the ordinary optics such as lenses and mirrors cannot be used for the x-ray imaging purpose as the index of refraction for x-rays has such values that it does not seem to be probable to get lenses for x-rays. The very first, technology for direct imaging the x-ray source is a pinhole camera. The pinholes are capable of imaging x-ray source with the additional appropriate filter to blind the pinhole camera for undesired radiations, and then function as a camera.

The pinhole-camera is the device that uses a tiny circular aperture to image an object into a dark box, permitting light rays to enter through this tiny aperture. The age of the pinhole camera technology is almost same as the parabolic mirror technology.
Alhazen (Ibn Al-Haytham) who is the author of the book of optics, around 1000AD, invented the first pinhole camera and named it as Camera Obscura. He explained the principle of pinhole camera and explained why the images were upside down. The first casual reference to the optic laws that made pinhole cameras possible, was observed and noted by Aristotle around 330 BC, who questioned why the sun could make a circular image when it shined through a square hole. The first photographic image was produced with a Camera Obscura by Joseph Nicéphore Niépce on the summer of 1827. Prior to Niepce the pinhole cameras were used for viewing or drawing purposes not for making photographs. Today pinholes cameras are obviously useful for imaging x-rays or particle streams where no lens materials are available. The advantages on the pinhole cameras are (i) offering complete freedom from linear-distortion, (ii) virtually infinite depth of focus and (iii) a very wide angular field.

In addition to x-ray source imaging purpose the aperture x-ray camera in XRIM is also used as an x-ray energy meter measure the energy of the x-ray source at particular wavelength range. In the chapter 4 the x-ray energy measurement procedure are presented.

The design of aperture x-ray camera in XRIM consists of a back illuminated Princeton Instrument (PI) CCD camera and Tantalum disc which has tripled apertures 100 μm, 25 μm, 50 μm for capturing simultaneously x-ray fluoresce channel images at two different wavelengths Xe L-shell and Xe M-shell from same x-ray source. The detail specifications of the PI along with the graph of quantum efficiency versus wavelength is given in the reference 41. The 2 μm thickness and 9.525 mm diameter circular Tantalum pinholes disc is a custom product of National Aperture Inc and the disc is mounted on an
aluminum cylinder to keep the apertures in specific distance from the CCD image-plane and perpendicular to the image-plane for producing desired magnification. The detail design of the triplet-aperture x-ray camera is presented in reference 41. The 25 μm, 50 μm apertures are blinded by using a 10 μm thick Beryllium filter foil for suppressing all radiation longer than $M$-shell and $L$-shell x-ray radiations and the 100 μm aperture is blinded by a additional 10 μm thick Titanium filter foil for suppressing all the radiation longer than $L$-shell x-ray radiation.

The triplet-aperture x-ray camera setup is viewing the target-area in the transverse direction with about 70 degrees of rotated angle from horizontal plane (x, y plane). Figure 2.19 shows the aperture camera setup along with all other diagnostics. The complete data sets of channel images captured by the triplet-aperture x-ray camera along with other diagnostics are presented in chapter 4. The data set recorded by the triplet-aperture x-ray camera shows that the spatial-resolution of the device is 120 μm.

### 2.3.3.2 Design of Von Hámos Spectrometers

Using the ordinary optics for imaging purposes in the x-ray wavelength is impossible as the ordinary optics offer poor reflectivity in the x-ray wavelengths regimes. On the other hand, the diffraction of x-rays by crystals, being equivalent to refraction, enables one to construct concave mirrors forming true x-ray images. It is practically possible that the concavely deformed suitable crystals could be use as an x-ray concave mirror for imaging purpose in the x-ray wavelength regime [43].

In 1916, M. Gouy pointed out that the cylindrically bent crystal has the propriety to give monochromatic stigmatic images of linear x-ray sources placed near to the
cylinder axis [44]. In 1932, von Hámos experimentally verified the idea and the scheme of the spectrometer were named after him. The von Hámos scheme had essentially forgotten for many years and it was reinvented in 1955. The primary role of the von Hámos spectrometers in XRIM is the spectroscopic study of the laser-matter interaction in addition to direct imaging of generated ultrabright x-ray source.

In a typical arrangement of von Hámos spectrometer scheme, for a spectrometric and imaging applications, the single-crystal is bent to form a cylindrical surface and the x-ray source and the image-plane of the detector lies on the cylinder axis. The x-rays from the x-ray source reach the cylindrically bended single-crystal and the crystal diffracts x-rays according to the Bragg formula presented below in the equation 2.1.

\[ 2d \sin \theta = n \lambda_x \]  
\[ (2.1) \]

Where \( n \) is an integer or order, \( \lambda_x \) is the wavelength of incident wave, \( d \) is the spacing between planes in the atomic lattice, and \( \theta \) is the angle between the incident ray and the scattering planes.

The diffracted x-ray from the cylindrically bend single-crystal are collected and focused to the axis of the cylinder to form a true image and spectral features. If the x-ray source is a monochromatic source, for every point of this source, there exists a corresponding point in the image-plane forming a monochromatic image. Beside the image, a whole spectrum of similar images is produced in the same image-plane if the source is not monochromatic source. The detail schematic diagram presented in figure 2.10 shows the spectrum of the non-monochromatic source produced in XRIM.

Two von Hámos spectrometers, presented in reference 41, were designed for the spectroscopic studies of Xe \textit{L-shell} x-ray emission during the laser-matter interactions.
The Xe $L$-shell von Håmos spectrometers are reconfigured, redesigned, and upgraded for the spectroscopic studies of Xe $M$-shell, Kr $L$-shell x-ray emissions in the experiments presented in this thesis. The crystal material of the von Håmos spectrometer of XRIM is ruby v-1 muscovite mica and the chemical composition is $\text{KAl}_3\text{Si}_3\text{O}_{10}(\text{OH})_2$. The mica is a commercial product of S & J trading Inc and the size of the mica is 4 cm x 6 cm x 0.002 cm. The detector is the PI back illuminated CCD camera. The size of the CCD chip of the PI camera is 3 cm x 3 cm; 2048 pixels x 2048 pixels, pixel size are 13.5 $\mu$m x 13.5 $\mu$m. The quantum-efficiency of the back illuminated PI camera is presented in reference 41 and efficiency of mica crystal at different orders is presented in reference 45.

The wavelength range of the spectrometer, size of the PI CCD camera chip, and other diagnostics configurations are the factors that determine the radius of the cylindrically deformed single-mica crystal and the size of the mica crystal. Simple calculations are performed prior to the device design to determine the size of the mica crystal, and the radius of the cylindrically deformed mica crystal for desired x-ray wavelength range. The atomic spacing ($d$) between two planes of mica crystal is 1.988 nm. The interest spectral regime of Xe $M$-Shell wavelength ($\lambda_x$) range 1.1 nm to 1.5 nm and the Bragg’s formula given in the above equation 2.4 restricts the order ($n$) of von Håmos spectrometer to $n = 1$ in the Xe $M$-shell wavelength regime. Since the angles of diffraction of the shorter wavelengths are smaller than the diffraction angles of longer wavelength, the longer wavelength falls on the image-plane closer to the source and the longer wavelengths falls in the image-plane farther to the source. The calculations showed that the diameter of the cylindrically deformed mica crystal of von Håmos spectrometer of XRIM should be at least 5.334 cm (2.1 inches) and angular-aperture (or
acceptance angle) of the cylindrically deformed mica crystal should be 129.25 degrees, and dimension of the mica crystal is 4 cm x 6 cm. A 5.334 cm diameter aluminum semi cylinder mandrel is machined and the mica crystal spread over the inside side surface of the semi cylinder as shown above figure 2.10.

The necessary conditions for the successful of operation of the von Hámos spectrometer is that the source and the image-plane of the detector should be lined up on the axis of the cylindrically deformed crystal. For aligning single mica crystal of the von Hámos spectrometer to satisfy all the necessary-condition mentioned above, the mica crystal holder is mounted on the PI camera with help of a Newport corporation linear-stage. The linear-stage (Part No: 9062-X-P) moves the mica-holder relative to the PI CCD camera-chip and perfectly aligns the CCD chip image-plane to the axis of the mica crystal. Further, the design have a removable alignment needle, which points at the spatial-location of the ultrabright x-ray source and runs through the image-plane of the CCD camera chip at the mica alignment position. Figure 2.10 below shows the design.
The wavelength range of the Xe \textit{M-shell} von Hámos spectrometer scheme is 1.1 nm to 1.5 nm. At the first order ($n=1$), according to Bragg’s formula, diffraction of the lowest extreme wavelength 1.1 nm and the diffraction of the upper extreme wavelength 1.45 nm produce 33.65 and 49.08 degrees diffraction angles. The Xe \textit{L-shell} x-rays lower-bound 0.22 nm and the upper-bound 0.3 nm produce the same diffraction angles 33.65 degrees and 449.08 degrees. Therefore, the design of the Xe \textit{M-shell} von Hámos spectrometer has an extra advantage of using it as fifth-order ($n=5$) Xe \textit{L-shell} von Hámos spectrometer with an introduction of appropriate filter to suppress any radiation longer than Xe \textit{L-shell} x-rays. It does not require any extra other mechanical modifications. Further advantage of the von Hámos spectrometers includes

- High spectral resolution, $\lambda/\delta\lambda \sim 1000$-2000
• One or two-dimensional direct imaging of the source
• High spatial resolution, \(\sim 50 \mu m\)
• High efficiency in a wide spectral range
• High sensitivity, focused intensities are \(10^2 - 10^3\) times higher compared with flat crystal spectrometers
• Mosaic focusing in the dispersion direction; Mosaic crystals provide high integrated reflectivity without sacrificing spectral resolution
• Very compact design
• Depth of field is small

We use two von Hámos spectrometer (one axial and one transverse) to monitor target-area. The basic designs of both spectrometers are same but the alignments of the spectrometers with respect to the x-ray channels are different in order to collect different type of spectroscopes information. The design of axial-spectrometer and transverse-spectrometer are presented separately in the following sections.

2.3.3.2.1 Design of Axial-von Hámos Single-crystal Spectrometer

The axial-von Hámos spectrometer in the XRIM experimental setup is aligned the crystal –axis of the spectrometer to make a Bragg angle with the propagation direction of the pump-laser. A main goal of the research presented in this thesis is to, experimentally, determine whether the realized x-ray source behaves as a laser. For the purpose of determine lasing, we choose monochromaticity, and collimation of the realized x-ray source as measurement of the axial-von Hámos spectrometer.
In the axial-spectrometer, the generated ultrabright x-ray source satisfying Bragg’s condition when it diffract from mica crystal could be used to determine whether the wavelength of the realized x-ray source is monochromatic. In the experiments for generation of ultrabright x-ray source, the directed x-ray radiations are believed to be guided and channeled by the pump-laser (experimental evident are presented in the 4.5 of chapter 4). The channeled x-ray radiations will be traveling in the direction of pump-laser with definite direction. If the lasing actions take place in the experiments, the dominating wavelength produced by the x-ray source will be a single wavelength $\lambda_x$. The single x-ray lasing wavelength $\lambda_x$ will travel forward-direction and diffract on the mica crystal immediately. The mica crystal could be aligned such a way to satisfy the Bragg’s condition for that particular single-lasing wavelength $\lambda_x$. The diffracted x-rays of this particular single-lasing wavelength $\lambda_x$ from the mica crystal will fall on a specific spatial location on the image-plane of the PI detector. If the source is produce dominated monochromatic x-ray radiation, a simple rotation of the mica crystal with an angle more than Rock curve about the x-ray source will violate the Bragg’s condition for the single lasing wavelength $\lambda_x$. The result of vanishing the signal at the particular spatial position of the detector image-plane will confirm whether the source behaves like a monochromatic source.

The axial-von Hámos spectrometer is an imaging device that can image a propagating beam at various axial-positions by translating the axial-spectrometer along the beam. If the size of the beam is same at the various positions of the propagating beam then it can conclude that the realized x-ray source has a laser like properties of collimation. In addition to these applications, given above, the axial-spectrometer is
capable of measuring experimentally, the brightness of the realized x-ray source. A theory for the brightness measurement is presented in the chapter 4 along with results.

A rotational motion of the axial-spectrometer mica crystal is the key to use the device for the applications mentioned above. However, the rotational motion should be performed extra carefully as it might cause to violate the fundamental operational conditions of the von Hámos spectrometers (the axis of the cylindrically deformed mica crystal should go through the x-ray source and the image-plane of the CCD chip). Therefore, the rotational motion of axial-spectrometer should be performed in such a way that the spectrometer keeps the x-ray source and the CCD image-plane at the axis of the cylindrically bend mica crystal. Crystallographers faces same scenario and they have resolved the problems with a goniometer. A goniometer is the device that rotates the mica crystal to any desired angle about the x-ray source. Such rotation will keep the x-ray source and the image-plane of the CCD in the axis of the cylindrically deformed mica crystal. From the facts above it is clear that the goniometer will be a potential candidate to perform the rotational motion of the mica crystal without violating the fundamental operational conditions of Hámos spectrometers.

The required angle of rotation of the mica crystal for satisfying the Bragg’s condition of the center Xe \textit{M-Shell} wavelength 1.3 nm is 41 degrees and the radius of the goniometer should be at least 20 cm radius for mechanically mounting the axial-von Hámos spectrometer as shown in figure 2.11. Designing or procuring a goniometer with the specifications given above was a challenging task. A goniometer with 20 degrees range of rotation angle (minimum incremental rotation is 0.5 micro-radians), and 20 cm radius procured from Huber Diffraktions Technik GmbH (part number: 5202.50) for the
axial-spectrometer construction. In order to increase the rotation range to 41 degrees a wedge is designed and mounted off-centered, as shown in figure 2.11, on the circle segment of the goniometer. An aluminum mirror is mounted on the wedge, as shown in figure 2.11, for the purpose of initial alignment of the spectrometer. Aligning the x-ray source to the center of goniometer is the key for the successful operation of the goniometer and the axial-spectrometer. An alignment needle, which could be mounted on the wedge, was designed to locate the center of the goniometer at the x-ray source. The alignment needle and goniometer are carefully designed such a way that the alignment needle could be removed from the wedge without disturbing the alignment, once the x-ray source spatial-location is aligned to center of the goniometer. The goniometer is mounted on a three-axes positioner such a way to move it in x, y, z linear-directions. The resolution of the positioner is 0.5 micrometers. In addition to the four-axes goniometer, a two axes positioner is built for aligning the x-ray source location (end-point of the goniometer alignment tool) to the axial-von Hámos spectrometers (end-point of the spectrometer alignment tool). The axial-spectrometer detector PI CCD chip is covered with the 10 µm Beryllium for suppressing the detector for all the radiations shorter than Xe M-shell x-rays. The detail design is presented in figure 2.11 below and the alignment procedures are following it.
2.3.3.2.1 Alignment Procedure of the Axial-spectrometer

In this section, alignment procedure for aligning the axial-spectrometer to 44 degrees Bragg angle at 1.38 nm is presented. The first step of the alignment procedure is to align the mica crystal of the axial-spectrometer to 44 degrees Bragg’s angle with respect to the direction of propagating pump-laser beam. Before the start of the alignment procedure, it is important to define the direction of the propagating pump-laser beam and the spatial location of the x-ray source. The following pre-alignment setup was design and built to align the axial-spectrometer with precision.

As a first step, the sonic-nozzle targets is installed on DGSN, and align the pump-laser focused-spot to the crosshairs of the sonic-nozzle target. Then the sonic-nozzle target is rotated to face the red laser-diode-beam and aligned it to the crosshairs. These two separate alignments guarantee that the pump-laser beam focused-spot is in alignment.
with the crosshairs of the sonic-nozzle holder. In other words, as shown in figure 2.11 the pump-laser focused-spot and center of sonic-nozzle orifice are at the origin of the virtual Cartesian coordinate system for the D chamber.

A flat-aluminum-mirror is mounted to a rotational-stage such a way that the flat surface of the mirror is lineup with the center of the rotational-stage. The center of the rotational-stage is lined up to the pump-laser focused-spot. The rotational-stage is then rotated such a way the front surface of the aluminum-mirror is normal to the red laser-diode. The back-reflected red laser-diode beam is then aligned to coincide with the incident-red laser-diode-beam by tip and tilt action of the aluminum-mirror holder. After aligned the aluminum-mirror the reflecting surface is now set parallel to the direction of the pump-laser beam. Next, rotational stage is rotated by 23 degrees (clock-wise) to send the reflected red-beam at 44 degrees to the pump-laser beam direction as shown in figure 2.12. At the end of these pre-alignments, the angle between the reflected red laser-diode beam from the aluminum-mirror and the pump-laser direction of the propagating is 44 degrees. Now the red laser-diode beam (which is making 44 degrees Bragg angle with pump-laser) is used to align the axial-spectrometer.
The step by step alignment procure for the axial-spectrometer are presented below

i. Install the goniometer positioner such a way the axes of positioner are parallel to the axes of the virtual Cartesian coordinate system. Incident red laser-diode beam on goniometer aluminum-mirror and the back-reflected laser beam coincide to with the incident beam.

ii. Install the alignment needle of the goniometer and install the sonic-nozzle target. Move the positioner of the goniometer to align crosshairs of the sonic-nozzle target to end-point of the alignment needle. The end-point of the needle is now at spatial-position of the focus of the pump-laser beam and the center of the goniometer.
iii. Remove the alignment needle, and make sure the incident diode red laser beam lies on the aluminum-mirror of the goniometer, and then ensure the back-reflected and incident red laser-diode beams are still coincide to each other. If the reflected red laser-diode beam is not coincident with the incident red laser-beam repeat, the procedures ii and iii until the back-reflected-beam is coincide with the incident-beam and the end-point of the alignment-needle matches with the crosshairs of the sonic-nozzle target.

iv. Remove the alignment needle and install the axial-von Hámos spectrometer along with the two-axes positioner and then install the alignment-needle of the spectrometer.

v. Check the end-point of the alignment needle of the axial-von Hámos spectrometer matches with crosshairs of the sonic-nozzle target. If they are not matching, move the two axes-positioners of the axial-von Hámos spectrometer and match the end-point of the alignment needle to the crosshairs of the sonic-nozzle target. At the end of the alignment, the x-ray source and CCD image-plane are now lined-up with the axis of the cylindrically deformed mica crystal as shown in the above figure 2.10.

vi. During the experiment to generate the ultrabright x-ray source, monitor the line thickness of the spectrum image of the axial-von Hámos spectrometer. As explained earlier a tiny linear-stage attached to the axial-spectrometer, which moves the semi cylinder of the mica crystal holder toward the axis and away from the axis of cylindrically deformed mica crystal. Move this tiny linear-stage until the spectrometer produces very sharp or narrow spectra line-image. At this
position, the axis of the axial-von Hámó spectrometer is in coincidence with the line, which connects the x-ray source and the image-plane of the PI CCD camera, as shown in the figure 2.10. This completes the alignment procedure for the axial-von Hámó spectrometer, which is necessary to operate the von Hámó spectrometers; the source and the image-plane, should be on the axis of the von Hámó spectrometers.

2.3.3.2.1.2 Von Hámó Spectrometers Calibration Estimation

The von Hámó spectrometers, like other spectrometers need to be calibrated before use for the spectroscopic applications. In XRIM, instead of calibrating the spectrometer the spectra collected by the spectrometers are performed to calibration estimation after experiments.

The calibration estimation is associated with a spectrum recorded by a flat-crystal spectrometer. The detail specifications of flat-crystal spectrometer are presented in reference 38. The wavelength range of the flat-crystal spectrometer is 0.85 nm to 1.5 nm. The diffraction crystal is KAP (Potassium Acid Phthalate) and the 2d spacing of the KAP is 2.6634 nm. The detector of the spectrometer was Kodak 101 films. In the spectrometer design, the x-rays were entering through a slit with slit-width 2 mm and the spectral resolution of the spectrometer was about 0.001 nm. The spectra taken with the flat-crystal spectrometer were generally intergraded over 360 to 720 laser-shots. A sample typical spectrum recorded by the spectrometer for studying Xe \textit{M-shell} and Kr \textit{L-shell} x-ray radiation are presented below in figure 2.13. The complete details of the spectra are presented in references 28, 46, 47, and 48.
Experiments where the flat-crystal spectrometer used as spectrometer is similar to the experiments where von Hámos spectrometer is used as a spectrometer to record emission spectral features. Further, both spectrometers are working based on similar principles. Therefore, fingerprints of emission-lines of spectra recorded by both spectrometers must be identical.

First of all, CCD pixel-voltage versus pixel-number graph plotted using IDL programming. In the graph, prominent spectral features are identified and corresponding pixel numbers are extracted. In the Xe spectra, the emission lines (i) \( \text{Sf}_27: \text{Xe}^{27+}; 3d^84s \leftrightarrow 3d^74s4f \) (ii) \( \text{Sf}_25: \text{Xe}^{25+}; 3d^{10}4s \leftrightarrow 3d^94s^24p4f \), and (iii) \( \text{Sf}_24: \text{Xe}^{25+}; 3d^{10}4s^2 \leftrightarrow 3d^94s4f \) are identified to use for calibration estimation (shown in figure 2.13 (a)). In the Kr spectra, the emission lines (i) K: \( \text{Kr}^{26+}; 3d \rightarrow 2p \), (ii) L: \( \text{Kr}^{26+}; 3d \rightarrow 2p \), and (iii) R: \( \text{Kr}^{25+}; 3s \rightarrow 2p \) are identified to use for calibration estimation (shown in figure 2.13 (b)).

An expression presented below in the equation 2.2 is used for calibration estimation.

\[
\lambda_{\text{lower}} + B \times \text{PixelNumber} \times \sin \theta_\lambda = \lambda \quad (2.2)
\]

Where \( \lambda_{\text{lower}} \) is the lower-limit of the spectra, \( B \) is a constant (label in this thesis as calibration constant), \( \text{PixelNumber} \) is the pixel number corresponding to an identified feature (could be evaluated from pixel-voltage versus pixel-number graph), \( \theta_\lambda \) is Bragg angle for the identified feature with wavelength \( \lambda \).

In the above equation 2.2, the flat-crystal spectra gives wavelength (\( \lambda \)) value for an identified features. Bragg angles (\( \theta_\lambda \)) corresponding to wavelength (\( \lambda \)) could be calculated using equation 2.1 and order of von Hámos spectrometer (\( n = 1 \) for Xe and \( n = 2 \) for Kr). The \( \text{PixelNumber} \) corresponding to an identified features could be exacted from the above mentioned pixel-voltage versus pixel number graph. Therefore, using the
equation 2.2 and 2 different identified features one could estimate the values for lower-limit spectra wavelength $\lambda_{lower}$ and spectrum calibration constant $B$. Once evaluated $\lambda_{lower}$ and $B$ values, using the equation 2.2 it is possible to estimate wavelengths corresponding to every pixel-number of spectra recorded by PI CCD camera. As a final step, pixel row going though image captured by von Hámos spectrometer is extracted using IDL programming and CCD pixel-voltage versus calibrated wavelength spectra plotted. Two sample spectra, those are processed through calibration estimation, are presented below in figures 2.13(a) (for Xe) and 2.13(b) (for Kr).

Even though the two figures 2.13(a) and 2.13(b) and flat-crystal spectra are in very good agreement, the calibration estimation introduces some errors. The value for the error is $\pm 0.005$ nm. Chapter 4, results and discussion presents calibrated Xe and Kr spectra and discussing physics behind the emission-lines.
Figure 2.13: (a): Von Hámos spectrometers calibration estimation for Xe
M-shell. The top picture is the spectrum collected by flat-crystal spectrometer. The bottom one is collected by von Hámos spectrometers.
2.3.3.2.2 Design of Transverse-von Hámos Spectrometer

The axial and transverse-von Hámos spectrometer have same basic design of a cylindrically-deformed mica-crystal together with a PI CCD camera setup, as presented at the beginning of above section. The different alignment setups and different positioner
setup are the only difference between the axial and transverse-spectrometers. The following paragraph discusses the design of alignment setup while the next paragraph discusses the design of positioner for the transverse-spectrometer.

The transverse-von Hámos spectrometer is aligned to the transverse direction to the direction of pump-laser-beam. The transverse-von Hámos spectrometer is designed to record simultaneously two separate x-ray spectra, one from the entrance-zone of the x-ray channel and the other one from exit-zone of the x-ray channel. In order to image the entrance-zone and exit-zone x-ray channels simultaneously, the von Hámos spectrometer alignment should satisfy two conditions. The first condition is that the axis of the von Hámos spectrometer must be aligned perpendicular to the propagating x-ray beam. The second conditions is that the plane, which is perpendicular to the CCD image-plane, which runs through spectrometer-axis must be perpendicular to the propagating x-ray beam (it is y, z plane according the figure 2.12). In the experimental setup of XRIM, the axis of the von Hámos spectrometer is rotated 60 degrees from the horizontal plane (x, y plane according to the figure 2.12) of D chamber without violating the two above conditions for imaging of the entrance and the exit-zone x-ray channels simultaneously.

The transverse-von Hámos spectrometer, unlike the axial-von Hámos spectrometer, does not require precise rotational alignment of the cylindrically deformed axis of the mica crystal. The axis could be rotated to any specific angle less than 60 degrees without violating the conditions presented above. Therefore, a very simple three-axes positioner could precisely align the transverse-spectrometer to its optimal alignment position. A three-axes positioner was designed and built, using Thorlabs Inc vacuum compatible linear stages (part number: PT1-Z8). The axes of the positioner are aligned
along the x, y, z direction. The actuators of the positioner are vacuum compatible commercial Thorlabs Inc product (part No: Z825B). The specifications of the positioner are 0.05 µm minimum achievable incremental movements, 0.2 µm minimum repeatable incremental movements and less than 8 µm backlash. Figure 2.14 below shows the transverse-von Hámos spectrometer the design of the three-axes positioner.

Figure 2.14: Schematic diagram of the transverse-von Hámos single-crystal spectrometer.

2.3.3.2.2.1 Alignment Procedure of Transverse-von Hámos Spectrometer

The transverse-von Hámos spectrometer captures x-ray channel image from transverse direction. The basic alignment procedures of the transverse-spectrometer are the alignment of the spectrometer-axis to the x-ray source, such a way that the alignments satisfy the fundamental operational conditions of the von Hámos spectrometer (x-ray source and CCD image-plane lies in the axis of the single-mica crystal). The transverse-spectrometer does not require precise x-direction alignment. Any misalignment in the x-
direction will only lead to a shift on the image-plane of the PI CCD camera, and it will not sacrifice any spectral information about the x-ray source. The systematic alignment procedure of the transverse-spectrometer is given below.

i. Install the sonic-nozzle target on the DGSN to define the x-ray source position, as well as center of the sonic-nozzle orifice.

ii. Install the alignment needle of the spectrometer that defines the axis of the spectrometer and the x-ray source point.

iii. Align the end-point of the alignment needle to the crosshairs of the sonic-nozzle target using the transverse-spectrometer positioners. At the end of the alignment, the x-ray source and the CCD image-plane lies on a line that is parallel to the axis of the spectrometer and satisfies the operation conditions for the transverse-von Hámos spectrometer.

iv. Remove the alignment needle. The tiny linear-stage of the spectrometer allow movement of the axis of the cylindrically-bend mica crystal back-and-forth with respect to the image-plane of the PI CCD camera by moving the mica crystal-holder. During the experiments to generate ultrabright x-ray source, actuating the tiny stage, allow to produce narrow spectrum line image-width. At the end of the alignment, the axis of the cylindrically bend-mica-crystal of the spectrometer and the straight-line, which connects the x-ray source and CCD image-plane are now coincided and the transverse-spectrometer is aligned.

The calibration estimation procedure for the transverse-von Hámos spectrometer is similar to calibration procedure of the axial-von Hámos spectrometer presented in the above section 2.3.3.2.1.3.
2.3.3.3 Design of UV Thomson Scattering Optical Setup

Thomson Scattering (TS), a classical phenomenon, is a scattering of electromagnetic wave by free non-relativistic electrons. The electric (E) and magnetic (B) components of the incident wave (photon) accelerate the electrons which are produced during the laser matter interaction. The accelerated electrons, in turn, emit radiation and thus, the wave is scattered.

J. J. Thomson following his discovery of electrons (1897) described the theory of Thomson scattering at the beginning of the 20th century (1906). Thomson scattering is a widely adopted phenomenon in plasma physics since the 1970's. TS has been widely used as a standard tool to detect high-temperature plasmas inside Tokamak reactors, and in other plasma experiments. In the XRIM laboratory TS diagnostics provides detailed information, such as distribution of produced electrons in the laser-matter interactions, density of electrons, the temperature of the electrons, the amount of pump-laser energy absorbed during x-ray generation, and self-channeling information to understand the physics behind the experiments that generate ultrabright x-ray sources. The detailed theory of the TS is presented in the chapter 4 along with the results. In this section, the diagnostic setup design and engineering, alignment procedures are presented.

TS diagnostic setup for relay imaging the scattered light from free electrons is similar to the focused-spot monitor optical-setup. The TS diagnostic setup consists of a micro-spot focusing-objective this mounted on a six-axis positioner that produce an aberrations-free magnified TS relay-image on the image-plane of a CCD camera that is mounted on a single-axis positioner. Micro-Spot UV focusing-objective is a commercial product (LMU-3x-248) of Thorlabs Inc. The details of the objective are (i) three times
image magnification, (ii) a 49 mm working distance and (iii) a 0.08 NA. The CCD camera is PI front-illuminated camera. The quantum-efficiency graph is presented in the appendix 2.6. The information of the PI CCD chip are 1340 x 1300 number of pixels, a 20 µm size square-pixels, and 2.75 x 2.75 cm CCD chip size.

The five-axes positioner for micro-spot focusing-objective is built by using three commercial linear-stages of Newport corporation (part number: 462-XYZ-LH-SD) and a commercial tip and tilt kinematic-optical-platform of Thorlabs Inc (part number: KM100R). The PI camera is mounted on a linear-stage that is similar to the linear-stage of the transverse-von Hámos spectrometer.

The direction of linear-polarization of pump-laser beam inside the D chamber restricts the line-of-sight of the TS setup to the in transverse direction (y-axis) in the horizontal (xy plane) plane. Since there are already two diagnostics (a triplet-aperture x-ray camera and a transverse-spectrometer) aligned to view the target-area in the transverse-direction to the propagating pump-laser-beam, building a TS setup in the transverse direction on the horizontal plane is a challenging design. A very compact optical-setup is built without interfering with the other diagnostics to extract a TS line-of-sight in the horizontal transverse direction (y-axis). The compact optical-setup divert the TS line-of-sight to a location where the micro-spot focusing-objective positioner and the imaging CCD camera are find enough room to mount and to align the TS five-axes positioner and the PI camera setup. In the compact optical-setup a device that divert the TS line-of-sight is a small aluminum-square-mirror (Thorlabs Inc, part No: 452784). In the TS setup, the small aluminum-square-mirror first collects the TS scattered-light in the
horizontal-transverse direction, then divert the collected photons to micro-spot focusing-objective to image, and magnify the TS object. The magnified TS images are then sent to PI CCD camera with a periscope setup. The PI camera is blinded to other radiations than 248.6 nm wavelength with three interference filters. The interference filters are product of CVI melles Griot (part number: F10-248.0-4-25.0M) and transmit 12 percent of 248.6 nm wavelengths-light. The transmission-curve of the interference-filter is presented in the appendix 2.7. Figure 2.15 below shows the TS setup. Figure 2.17 shows the TS setup together with other diagnostic setups.

2.3.3.3.1 Thomson Scattering Alignment Procedure

Even though the optical-setup and the imaging principles for TS are almost similar to the focused-spot monitoring optical-setup and its imaging principles, the alignment procedures are very different to both optical-setups. In the focused-spot monitoring alignment procedures, it was possible to produce the focus-spot of the seeding-beam prior to begin the generation of ultrabright x-ray source experiments but in the TS it is not possible to produce TS image prior to begin the generation of ultrabright x-ray source experiments. Therefore, the alignment of the TS imaging system requires a real-object mounted inside D chamber for the TS setup alignments prior to begin the experiments. The object used prior to the experiments is a calibrated target such as, Ronchi target mounted such a way that the vertical-symmetrical object-plane of the TS (which only takes place during experiments. Here it is assumed that symmetrical vertical-plane of TS is in the x, z plane and it is going though center nozzle orifice) of object will coincide to the object-plane of the target. Once the alignment is completed, this object
removed. Figure 2.15 below shows the experimental-setup to align TS diagnostics-setup prior to begin the main experiments.

![Figure 2.15: Schematic diagram of TS setup.](image)

Keeping the magnification of the micro-spot focusing-objective to the designed magnification (= 3) during the alignment of the TS is one of the major controlling factors required to produce aberration-free, magnified TS images by the micro-spot focusing-objective. Aligning the TS micro-spot focusing-objective to its designed magnification also requires a calibrated target mounted in a vertical-plane through which the symmetrical vertical plane of the TS object will fall during the experiment for the generation of ultrabright x-ray source. The figure 2.15 above shows the setup for tuning the magnification to designed magnification.

The TS setup alignment procedures consist three steps (I) aligning the TS optical-setup for achieving aberration-free imaging-system (II) Setting the magnification of the micro-spot focusing-objective to its designed magnification (3) (III) experimentally
determining resolution of the TS imaging-system. The systematic alignment procedure is presented below.

I. **TS alignment procedure for producing aberration-free relay-images**

The micro-spot focusing-objective, which magnify and image the scattered radiation from free-electrons is similar to other imaging-systems and requires six-independent alignments degrees-of-freedom and proper systematic alignment procedure for producing aberration-free images. The systematic alignment procedures are presented below.

I.I. Remove micro-spot focusing-objective from the TS positioner and turn on the red laser-diode. The red laser-diode-beam goes through center of the DGSN and reflects in the small rectangular-aluminum-mirror and then it makes two reflections in the two periscope mirrors (shown in the figure 2.15).

I.II. Locate the red laser-diode-beam after it is reflected in the second-mirror of the periscope and then setup crosshairs at the center of the beam.

I.III. Install micro-spot focusing-objective on the six-axes positioner. If the beam is not aligned on the crosshairs move the objective-positioner until center of the red laser-diode-beam hits the crosshairs. Remove micro-spot focusing-objective and make sure the red laser-diode-beam is aligned to the crosshairs.

At the end of these three alignment steps, the micro-spot focusing-objective is producing aberration-free images with the unknown magnification. Since the objective is designed for three-time magnification, setting the magnification of the micro-spot focusing-objective to three-time magnification drastically improves the image quality.
The alignment procedure for setting up the magnification of the micro-spot focusing-objective to manufacture magnification (= 3) are presented below.

II. Setting the magnification of the TS objective

A combination of image and object distances, similar to that of an ordinary lens, determines the magnification of the micro-spot focusing-objective. The magnification alignment procedures involve with successive adjustments methods. Every time when the object distance and image distances are changed to alter the magnification of the TS setup to the manufacturer magnification value, the imaging camera has to take pictures of the magnified image of the calibrated target, then calculate the magnification of the TS setup at this particular image and object distances. Because of various reasons, the PI camera is not user friendly for the purpose of successive adjustment procedures. Therefore, tentative magnification tuning procedures were performed with the help of Lumenera camera (Part number: LW-235-M). The pixel size of the Lumenera camera is 4.5 x 4.5 µm and the size of the CCD chip is 1 x 1 cm. Further, the camera is trigger-able and user friendly in this application. After the tentative alignment is achieved, the final magnification was achieved using the PI camera as explained below in the procedure. The procedure for adjust the magnification of micro-spot focusing-objective to manufacturer magnification are presented below.

II.I. Mount a plastic transparent metric ruler vertically at the center of nozzle. The ruler is aligned such a way that the front-plane of the ruler and the vertical center-plane of the propagating pump-laser beam coincides.
II.II. Remove PI camera and install a millimeter scaled inspection paper where the PI camera chip is located. The image of the rule lines will be on the inspection card when the red laser-diode is ON.

II.III. Move the optical-setup of the six-axes positioner and the periscope system together, back-and-forth, along the beam reflected from square aluminum-mirror until magnified two-millimeter division image-lines matches to the six-millimeter division-lines of the scaled inspection-paper. Now the TS setup is tentatively tuned for magnification of three.

II.IV. Repeat the procedures from I.I to I.III and make sure the micro-spot focusing-objective is aligned to produce an aberration-free image.

II.V. Install the Lumenera CCD camera where the image-plane of the CCD camera and the font-plane of the inspection paper coincide.

II.VI. Replace the plastic millimeter ruler with calibrated fused-silica Ronchi slide-target. The calibrated fused-silica Ronchi-slide-target is a commercial product of Edmund optics (part number: NT57-886). In this target, the distance between two lines is 50 µm.

II.VII. Install a collimated rare Hg-gas lamp such a way the collimated UV photons of the lamp are propagating in the direction of the red laser-diode-beam (y-axis). The rare Hg lamp is commercial product of Ultra Violet Products (UVP) LLC (Part number: 90-0017-01). The dominant wavelength of the Hg gas lamp is 254.3 nm.

II.VIII. Move slightly the Lumenera CCD camera back-and-forth, by using the CCD camera linear-stage, until the micro-spot focusing-objective
produces its sharpest images on the CCD camera chip. Snap a picture of the magnified image of the calibrated target.

II.IX. Use the IDL software and count how many lines lie in between one hundred CCD pixels. The magnification can be calculated from the formula; Magnification of the TS setup = (100 x pixel size of the CCD)/(Number of lines -1).

II.X. If the magnification is not exactly three, move the micro-spot focusing-objective to produce designer magnification (3) and repeat the procedure II.VII and II.IX.

II.XI. Now replace the Lumenera camera with PI camera such a way image-planes of both cameras coincide.

II.XII. If the magnified calibrated-target image on the CCD camera is not sharp, move the PI camera back and forth by using the PI camera linear-stage until it produces sharp image.

II.XIII. Take a picture of the calibrated-target and repeat the procedure II.IX and calculate the magnification of the TS setup. At this position, the magnification of the micro-spot focusing-objective should be very close to the manufacture magnification. If the magnification is not exactly same to the manufacturer magnification, repeat the procedures from II.X and then II.XII and II.XIII until the TS produces manufacturer magnification. Figure 2.16, below, shows the sample picture of magnified calibrated-target snapped by the Lumenera and PI camera respectively.
As shown in appendix 2.8, the effective focal-length of the micro-spot focusing-objective is depend on wavelength. In the experiments for generation of ultrabright x-ray source the TS object is produced by 248.6 nm wavelength radiations and the alignment of micro-spot focusing-objective magnification to manufacturer magnification performed using a 254.3 nm wavelength (UV) Hg gas lamp. Therefore, the micro-spot focusing-objective requires a correction for effective focal length at 248.6 nm. The correction for the effective focal length calculated from the effective focal length graph presented in the appendix 2.8 and corresponding wavelengths. The effective focal-length correction at 248.6 nm is – 300 µm. Therefore, the micro-spot focusing-objective should be moved 300 µm towards the TS object in order to produce 3 times magnified aberration-free images at 248.6 nm. Moving micro-spot focusing-objective for the correction changes the object-distance, image-distances, and this movement slightly changes magnification and the TS image will be blurry. Moving the PI camera for a sharper image during the experiments for generation of ultrabright x-ray sources will tune the magnification of the
micro-spot focusing-objective to designed magnification by changing the image distance. At this PI CCD camera position, the TS optical-setup is aberration-free and produces three-times magnified-images on the image-plane of the PI CCD camera chip.

### 2.3.3.3.2 Experimentally Determining Spatial Resolution of the TS Imaging System

In order to determine the resolution of the TS diagnostic setup a calibrated fused-silica Ronchi slide-target is replaced by a USAF resolution target. The resolution USAF target is a commercial product of Thorlabs (part number: R3L3S1N). As a first step, the PI CCD camera was replaced with a Lumenera camera (LM-150-M) and the resolution of the TS setup with Lumenera camera was determined experimentally. The resolution USAF image captured by the pair of micro-spot focusing-objective and Lumenera CCD camera is given below in the above figure 2.16. Finally, the Lumenera CCD was replaced with the PI CCD camera and the resolution of the TS was experimentally determined. The resolution USAF image captured by the pair of micro-spot focusing-objective and the PI CCD camera is presented above in the figure 2.16.

The resolution of the experimental setup depends on the pixel size of the CCD camera and the wavelength of the source used for the imaging. The USAF resolution image presented in the figure 2.16(b) is captured from a scenario, which is almost similar to the experimental situation for generation of ultrabright experiment. The only difference is that the collimated UV Hg gas lamp source for imaging purpose has different wavelength than the real situation. The wavelength of the collimated UV rare Hg gas-lamp source is higher than the wavelength of the TS source during the experiment for generation of ultrabright x-ray source. The resolution of typical imaging systems, similar
to the TS, depends on the source wavelength and when wavelength goes down the resolution will go up. Therefore, it is obvious that the resolution obtained from the figure 2.17.2 has positive error as it was experimentally determined using a higher wavelength source. In other words, the exact resolution of the TS setup is higher than the resolution presented the above figure 2.16. The resolution of the TS is greater than or equal to 36 lines per mm or 24.8 µm. However, the smallest image captured by TS optical-setup in the experiments presented in this thesis has size of 27 µm.

Figures 2.17, 2.18, and 2.19, presented below, are show diagnostic setup. Since the target-area is crowed with four different diagnostics and it is difficult to get a line-of-sight to show the entire diagnostics setup, the diagnostics setup is presented in three steps. The first step in figure 2.17 shows only alignment of axial-spectrometer and TS. The second step in the figure 2.17 shows axial and transverse-spectrometer alignments along with TS setup. The final third step shows the diagnostics alignment setup of XRIM for generation of ultrabright x-ray source.
Figure 2.17: Axial von Hámos Hamos crystal spectrometer and the TS setup

Figure 2.18: Axial, transverse-von Hámos Hamos crystal spectrometers and the TS setup.
The designed diagnostics are eyes of the XIRM experiments, which witness success of the experimental design. The design and engineering of the calibrated XIRM diagnostics and XIRM infrastructure produce a tremendous amount of data during the interaction of pump-laser with target-clusters. The rate of data production of the XIRM lab reached new level, and shows the success of design and engineering of the diagnostics and infrastructures. The following chapter 3, theoretical framework, is developing a tool to understand the experimental single-shot data recorded by the calibrated diagnostics and hence the physics behind the generation of laboratory-scale ultrabright x-ray source.
3. THEORETICAL FRAMEWORK

3.1 Introduction

Nowadays there are various theories and theoretical models are available to understand the physics behind the generation of laboratory-scale ultrabright kiloelectron Volts x-ray sources. Therefore, instead of developing new models and theories, essence of previously developed theories and models of XRIM are borrowed to explain physical observations of the ultrabright x-ray source experiments presented in this thesis. Particularly mathematical expressions describing physical parameters are borrowed from previously developed theories and applied in this thesis to estimate physical-quantities. Brief introductions to following sections are presented below.

Producing necessary pump-power for lasing action at the x-ray wavelength is one of the important keys for realizing x-ray laser at laboratory-scale. The section 3.2, estimating pump-laser intensity at target-medium, discusses the aptitude of pump-laser of XRIM at the target-medium. First, focal-spot waist-size at focal-plane is estimated at ideal conditions and then it is estimated in real experimental scenario. Section 3.2 ends with summarizing pump-laser parameters at focal-plane of the focal-spot where it interacts with target-atoms.

Like in the conventional lasers, selection of lasing-medium with suitable condition for lasing action is also an important factor for the success of generation of ultrabright x-ray sources at laboratory-scale. The Xe and Kr clusters produced by a sonic-nozzle are the lasing-medium in XRIM. In section 3.3, simple estimations are performed to determine target-parameters such as density and size of the target-clusters produced by
the sonic-nozzle (DGSN). Section 3.3 ends with summarizing target-clusters parameters in tabular format.

One of the fundamental conditions for success of lasing action is creating population inversion. Section 3.4, generation of ultrabright x-rays from laser-matter interactions, discusses selective ionizing mechanisms, which create population inversions to generate ultrabright keV x-rays. Section 3.4 begins with introducing Xe and Kr target-materials. It is then, extended to discuss various ionization mechanisms taking place during interaction of pump-laser with target-medium at previously estimated pump-laser intensity level. A theoretical model termed as intra-cluster-model sets constraints to both, the pump-laser intensity and target-cluster size, for successful generation of ultrabright x-ray sources. At the end of this section, a new ionization mechanism termed as dynamical orbital collapse is briefly discussed.

Directionality is one of the fundamental natures of lasers. In section 3.5, a mechanism, which channel the generated ultrabright x-ray radiations from selective ionization mechanisms are briefly discussed. Section 3.5 begins with defining x-ray channels and then discussing fundamental mechanism, which self-channel generated ultrabright x-rays. A mathematical expression describing conditions for stable self-channel is borrowed from theoretical works of XRIM and presented. The mathematical expression sets conditions for stabilities of x-ray self-channel and lays foundation for explaining experimental observations of different x-ray channels morphologies at the end of x-ray channels. A simple idea of diffractions in a finite-aperture is proposed to estimate x-ray channels diameters experimentally.
3.2 Estimating Pump-laser Intensity at Target Material

Estimating available pump-laser intensity at the target-material for the laser-matter interaction helps to understand physics involved in the generation of ultrabright x-ray sources experiments. The estimation of pump-laser intensity depends on experimental measurement of laser-power and focal-spot beam waist-size. Measuring experimentally, high intensity laser-beam focal-spot waist is a time consuming and a challenging experimental problem. Therefore, in XRIM during the daily experiments low intensity seed-laser beam focal-spot waist-sizes are measured prior to begin real experiments for generation of ultrabright x-ray source. This thesis presents a method to estimate the high-intensity focal-spot beam waist-size and the available intensity at the target-medium. The estimation of available pump-laser intensity at target-medium in the real experimental conditions begins with the estimation of seed-beam focal-spot waist-size presented below in following section.

3.2.1 Estimating Seed Beam Focal Spot Waist Size

Diagnostics beam-profile experiments performed on the Hurricane laser-beam shows that the beam emitted from the Hurricane laser has a Gaussian transverse irradiance-profile that varies radially from the propagating-axis. Mathematically, the irradiance-profile of the focal-spot produced by the off-axis parabolic mirrors is a Fourier transform of incident beam-profile and Fourier transform of a Gaussian function results again in a Gaussian function. Therefore, if the seed-laser pulses are holding the Gaussian profile nature just before it reaches the Off-Axis Parabolic (OAP) mirrors, one could
expect the aberration free focal-spot produced by the OAP is also a Gaussian focus. The mathematical descriptions of the Gaussian focus are presented below.

The mathematical description of the propagating electromagnetic wave of laser-beam in time and space is given blow by a formula

\[ \nabla^2 u = \frac{1}{c^2} \frac{\partial^2 u}{\partial t^2} \]

(3.1)

Where \( c \) represents the speed of the propagating wave and \( \nabla^2 \) is the Laplacian-operator defined in rectangular coordinates by \( \nabla^2 u = \frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} + \frac{\partial^2 u}{\partial z^2} \).

The time variation in the filed is sinusoidal and the laser produces monochromatic wave, the solution of the equation 3.1 is in the form of \( u(r,t) = u_0(r) e^{-i\omega t} \). Substituting the solution in the equation 3.1 leads to the time-independent reduced wave equation or Helmholtz equation.

\[ \nabla^2 u_0 + k^2 u_0 = 0 \]

(3.2)

Where \( k \) is the optical wave-number related to the optical-wavelength \( \lambda \) by \( k = \omega / c \)

In the optical-wave propagation, the free-space optical-field at any point along the propagation path remains rotationally symmetric. Therefore, the optical-wave propagating in the \( z \) direction could be expressed in cylindrical coordinates as a function of \( r = \sqrt{x^2 + y^2} \) and \( z \) as presented below in equation 3.3.

\[
\frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial u_0}{\partial r} \right) + \frac{\partial^2 u_0}{\partial z^2} + k^2 u_0 = 0
\]

(3.3)

Substituting the solution \( u_0(r,z) = V(r,z) e^{ikz} \) in the equation 3.3 leads to
\[
\frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial V}{\partial r} \right) + \frac{\partial^2 V}{\partial z^2} + 2ik \frac{\partial V}{\partial z} = 0
\]  

(3.4)

The paraxial approximation is based on the notion that the propagation distance for an optical-wave along the \(z\)-axis is much greater than the transverse spreading of the wave. By using paraxial approximation, the wave equation is further simplified as presented below.

\[
\frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial V}{\partial r} \right) + 2ik \frac{\partial V}{\partial z} = 0
\]  

(3.5)

The solution for the above equation 3.5 could be sought through Huygens-Fresnel integral. The details of solving the paraxial approximated Helmholtz equation 3.5 is not presented in this thesis. The typical solution scalar-paraxial wave equation (Helmholtz equation) is a Gaussian beam, given by:

\[
u(r, z) = \frac{w_0}{w(z)} \exp \left( -\frac{r^2}{w^2(z)} \right) \exp \left( -i \frac{kz - \xi(z) - \frac{br^2}{2R(z)}}{2} \right)
\]  

(3.6)

In the equation 3.6 \(w_0\) is beam waist-size at \(z = 0\) location, where the field is down to \(1/e\) compare to on axis and 86 % of beam-power contained inside the beam-waist. The other parameters \(w(z), R(z), \) and \(\xi(z)\) are defined below

\[
\text{Beam-waist size } w(z) = w_0 \sqrt{1 + \left( \frac{z}{z_0} \right)^2}
\]  

(3.7)

\[
\text{Radius of curvature } R(z) = z_0 \sqrt{1 + \left( \frac{z}{z_0} \right)^2}
\]  

(3.8)

\[
\text{Gouy phase } \xi(z) = \tan^{-1}\left( \frac{z}{z_0} \right)
\]  

(3.9)
The parameters $z_0$ could be determined from the boundary conditions and known as Rayleigh range. The Rayleigh range $z_0$ is defined by the distance, which the beam travels from the beam-waist before the beam-diameter increases by $\sqrt{2}$, or before the beam-area doubles. Further, within the Rayleigh range points $z_0$ the beam spread is minimum and when the beam propagating out from Gaussian waist reaches the Rayleigh range point $z_0$ the beam begins to diverge significantly. The Rayleigh range marks the approximate dividing line between the near-filed or Fresnel and the far-filed or Fraunhofer regions for a beam propagating out from the Gaussian beam waist. Further, the Rayleigh distance $z_0$ is the axis location where the phase on the beam axis is retarded by $\frac{\pi}{4}$ relative to plane-wave and the intensity is dropped to half of axial-intensity at beam-waist. The mathematical expression for Rayleigh range is presented below in equation 3.10.

$$\text{Rayleigh range } z_0 = \frac{\pi w_0^2}{\lambda L}$$ (3.10)

The Gaussian beam parameters are shown, briefly, in figure 3.1.
After the Rayleigh range, the propagating beam from the Gaussian beam-waist diverges rapidly as shown in the above figure 3.1. The derivation for estimating the divergence angle is presented below. At an axial-position \( z \) of the propagating beam, where the value of \( z \) is very much greater than the Rayleigh range \( z_0 \), the first term of the equation 3.7 is negligible. Therefore, the equation 3.7 simplifies as shown below in equation 3.11.

\[
w(z) = w_0 \left( \frac{z}{z_0} \right) = 2z \tan \theta_0
\]

\[
\Rightarrow \tan \theta_0 = \frac{w(z)}{2z} \quad (3.11)
\]
At \( z = z_0 \), \( w(z_0) = 2w \), the equation 3.11 further simplifies to

\[
\Rightarrow \tan \theta_0 = \frac{w_0}{z_0}
\]

(3.12)

Substituting the expression for the \( z_0 \) presented in the equation 3.10 on the equation 3.12 leads to half divergence-angle presented below in equation 3.13

\[
\theta_{beam} = \theta_0 = \frac{\lambda_L}{\pi w_0}
\]

(3.13)

The above theoretical derivations are performed based on the assumption that the Gaussian beam-pulses emitted by the Hurricane laser are not truncated before it produces the focal-spot. The Hurricane laser seed-beam passes through 11 windows, four Excimer-amplifier gas-chambers filled with KrF* gas and four lenses and they reflect in 26 flat-mirrors, two diffraction Littrow gratings and three spherical-mirrors before it reaches the focal-plane. Therefore, the assumption, the seed-beam focal-spot irradiance-profile is a Gaussian distribution, might be a deceiving assumption. In order to verify the assumption, seed-beam focal-spot is experimentally measured and the transverse-irradiance-profile of the focal-spots was fitted to the Gaussian function as shown in figure 3.2.

In XRIM, prior to begin experiments for generation of ultrabright x-ray sources, the seed-laser pulses are focused to possible smallest focal-spot size. The image of seed-beam focused-spot is relay-imaged and captured using an optical-setup, which consists of micro-spot focusing-objective and a CCD camera. The seed-laser beam focused-spots parameters such as irradiance-profile of the focal-spot, the focal-spot waist size at the focal-plane, Rayleigh range, beam divergence angle, and depth of focus are estimated using a code written in IDL. Figures 3.2 (a) and 3.2 (b), presented below, show the 3-D
irradiance-profile of the seed-laser focal-spot. For these purposes, a row-vector, which goes through the maximum intensity pixel is extracted and plot against the pixel number. The plotted graph is then fitted with typical Gaussian function in order to verify the above assumption (seed-beam focal-spot irradiance-profile is a Gaussian distribution). The $1/e$ value of the Gaussian function is estimated and focal-spot waist-size at the $1/e$ value of the Gaussian function is determined as shown in figure 3.2 (a) presented below.

![Figure 3.2 (a)](image)

![Figure 3.2 (b)](image)

Figure 3.2: 20 times magnified focal-spot irradiance-profile. (a) Irradiance-profile distribution in along x, y-directions and data points are fitted to Gaussian function. (b) Focal-spot irradiance-profile plotted in contour, and shaded-surface.

The seed-beam focal-spot parameters are presented below.

- Focal-spot waist size $w_0$ is $\sim 2.5 \, \mu$m
- Rayleigh range $z_0$ is $79 \, \mu$m
- Half-divergence beam angle $\theta_{beam}$ is 1.8 degrees
The laser-beam after it is reflected from OAP converges to form a sharp focal-spot. The beam diameter just before it is reflected from the OAP is \( D \) and focal length of the OAP is \( f \). Since the OAP converging beams satisfies the condition \( z >> z_0 \) in the equation 3.7 first term could be negligible. The simplified equation is presented below in equation 3.14.

\[
w(z) = w_0 \frac{z}{z_0}
\]  

(3.14)

In the equation 3.14, the \( w(z) \) is equal to \( D/2 \) and the \( z \) is equal to \( f \). By introducing these parameters and using the equation 3.10 for Rayleigh range \( z_0 \) the modified version of the equation 3.14 is presented below.

\[
\frac{D}{2} = w_0 \frac{f}{\pi w_0^2} \frac{1}{\lambda_L}
\]

\[
\Rightarrow w_0 = \frac{2f\lambda_L}{\pi D}
\]

The theoretical Gaussian beam waist-size without including the \( 1/e \) criteria is given below in equation 3.15.

Theoretical focal-spot radius at the focal-plane \( r_{focus} = \frac{4f\lambda_L}{\pi D} \)  

(3.15)

In XRM, the wavelength \( \lambda_L \) is 248.6 nm, focal-length \( f \) of OAP is 203.2 mm, and the laser beam is 90 x 95 mm rectangular beam. If one assumes that the beam has an effective radius (\( D/2 \)) of 46.25 mm, the estimated theoretical focused-spot beam waist-size at the focal-plane \( w_0 \) is \( \sim 0.7 \) \( \mu \)m for the seed-beam parameters presented above and the NA is 0.22.
3.2.1.1 Beam Propagation Factor $M^2$

All the above estimations are performed based on the fundamental assumptions that the Hurricane laser is operating at extremely high TEM$_{00}$ single-mode quality. Since the multimode combinations can have a nearly perfect Gaussian, the Gaussian fit to the transverse-irradiance profile, presented in the figure 3.2, might provide false information about the operating mode of the laser. A dimensionless parameter known as laser propagation-factor ($M^2$) is defined, which quantitatively compares the propagation characteristics of the real beam to those of a pure TEM$_{00}$ Gaussian beam. The laser propagation factor in terms of the laser parameters is presented below in equation 3.16.

$$r_{\text{focus}} = M^2 \frac{4f_2L}{\pi D}$$  \hspace{1cm} (3.16)

The equation 3.16 shows that the focal-spot radius $r_{\text{focus}}$ is $M^2$ times larger than it would be for a pure TEM$_{00}$ Gaussian beam of the same input beam-size $D$. According to the definition of International Organization for Standardization (ISO), Geneva, Switzerland the beam-waists are measured in several locations after and before the focal-plane in order to experimentally measure $M^2$. Plotting the measured $1/e^2$ beam-waists against the distances from the focal-plane will lead to estimate the divergence-angle of the propagating beam, the focal-spot beam-waist, and the propagation factor ($M^2$). The estimation of $M^2$ will lead to determine how close the pump-laser beam is to TEM$_{00}$ Gaussian beam. The estimation of $M^2$ is not presented in this thesis but detailed estimation of $M^2$ of the XRIM pump-laser will be presented in a research paper along with other pump-laser parameters.
In the experiments for generation of ultrabright x-rays, energy of the pump-laser is roughly one thousand times higher than the seed-beam. Therefore, one could expect focal-spot qualities of the high power pump-laser pulses to be different from seed-beam focal-spot qualities. Measuring high intensity focal-spot waist-size using the same experimental setup designed for measuring seed-laser focal-spot waist-size demands attenuation of high intensity laser-beam by almost ten-thousand times. Therefore, estimating high intensity focal-spot waist-size requires a completely different experimental setup and approach. Although, in this thesis, two independent theoretical and experimental methods are proposed to estimate high power focal-spot waist-size, this thesis is mainly focusing on the experimental method to estimate high power focal-spot waist size.

3.2.2 Estimating High Intensity Focal Spot Waist Size

The seed-laser pulses have ~ 50 µJ average energy at the output of tripling-box. This energy is not enough to introduce reasonable amount of nonlinear effects to the propagating laser pulses from tripling-box to the target-medium. But in the real experimental scenario, about 20-30 mJ pre-amplified laser-pulses propagate from LLG 50 PRO to Prometheus and then final amplified laser-pulses with average energy of 250-400 mJ propagate through moderate vacuum (about 50 mtorr) from Prometheus output to the input of the experimental D chamber. The propagating high-power pump-laser pulses face various nonlinear effects during their propagation and alter quality of the focal-spot, as the focal-spot quality is Fourier transforms of pump-laser beam irradiance-profile just
before it reflects from OAP. Therefore, one could expect that the laser focal-spot quality in the real experimental scenario is different from the seed-beam focal-spot qualities.

Further, the DGSN’s solenoid puppet opens hundreds of microseconds prior to the pump-laser-pulses reach the focal-plane where it interacts with target-clusters. The target-gas molecules leave the DGSN orifice with a velocity equal to the speed of sound (346 m/s). Therefore, before laser-pulses reach the focal-plane, in hundreds of micro-seconds time window, the target-gas molecules spread into space in the order of centimeters length-scale and create a gas-column through which the high-power laser has to travel before it reaches focal-plane. This target-gas column not only absorbs the pump-laser energy before it interacts with target-clusters but also alters the irradiance-profiles by introducing nonlinear effects to the laser-pulses during their propagation through the target-gas column before it reaches focal-plane where it interacts with target-medium. One of the major nonlinear mechanisms, which alter high power laser-pulses during their propagation through medium, is self-focusing mechanism. A discussion is presented below to show how the self-focusing mechanism alters the irradiance-profile and focal-spot quality.

The refractive index \( n \) of a medium through which the high-intensity beam propagates could be expanded in terms of field-strength as \( n = n_0 + n_2 E^2 + \ldots \). Such medium is known as optical Kerr-medium. Consider a laser beam propagating with the irradiance-profile as shown in the figure 3.1 entering a optical Kerr-medium with index of refraction \( n = n_0 + \eta_2 I(r) \), where \( I(r) \) is the beam intensity distribution along the radial coordinate \( r \) and nonlinear refractive index \( \eta_2 > 0 \). The central part of the beam sees a larger \( n \) and therefore moves slower than the edge and the wave-front gets distorted. Since
the optical rays always propagate perpendicular to the wave front, the beam appears to focus by itself. However, in other hand beam with finite cross-section diffract and defocus. If the self-focusing effect and the diffraction effect are exactly same, they cancel each other and the beam would propagate without any change on its spatial-profile. This is the case of self-trapping, also known as the spatial soliton. However, the solution is metastable. A small increase or decrease of the beam intensity irradiance-profile would upset the balance for self-trapping and make the beam self-focus or diffract. Producing and transporting ideal homogeneous beam-profile without introducing any distortions is not practically possible. Therefore, self-focusing phenomena is an inevitable mechanism during the propagation of high-power lasers through medium.

The critical-power, the minimum laser-power required for self-focusing mechanism take place, is presented below by equation 3.17.

\[ p_{cr} = \frac{3.72 \lambda^2}{8 \pi n_0 n_2} \]  

(3.17)

A simple estimation for the critical power \( p_{cr} \) shows that the pump-laser power of the laser-beam inside the experimental-chamber is higher than the critical-power \( p_{cr} \). Therefore, one could expect strong self-focusing effects during the beam propagation. The effect of self-focusing could be divided into two. They are (i) whole beam self-focusing (ii) filamentation; if the pump-laser beam-power \( P_L \) is very much higher than the critical-power \( p_{cr} (P_L \gg \gg p_{cr}) \) the beams break into filaments each with critical-power \( p_{cr} \). Introducing any minor distortions into spatial-profile of the propagating high-power beam will trigger the filamentation mechanism and the beam propagating distance will accelerate the triggered filamentation mechanism.
It is clear from all of the facts above that the self-focusing mechanism alters the focal-spot quality by altering irradiance-profile of propagating beam. Therefore, it is important to take into account the effect of self-focusing during the estimation of the high-intensity focal-spot beam waist. A mathematical expression for the whole beam self-focusing action after the plane-wave of laser-pulses reflected from OAP is borrowed from reference 49 and presented below as equation 3.18.

\[
\theta_{in}z = \left( \frac{8n_r^2 P_L}{n_0^2 cr} \right) - r
\]

(3.18)

Where \(\theta_{in}\) is the initial converging-angle, \(r\) is the ray’s distance from the axis, \(c\) is velocity of light, and all other parameters are previously defined meanings.

The equation 3.18 shows that the beam path towards the focal-plane from the OAP not to follow straight-line but the hyperbola. Therefore, the focused beam never reaches a focus, but approaches the beam-axis asymptotically. By solving equation 3.18 for \(r\) and substituting values for all parameters will lead to estimate, theoretical focal-spot-waist size. However, in this thesis such estimations are not presented.

The high-power laser focal-spot waist-size is estimated from Thomson scattering experimental method. The physical phenomenon, scattering of pump-laser radiations from free electrons is known as Thomson Scattering (TS). The introduction to TS and TS experimental designs are presented in the chapter 2. The designed TS diagnostics is an instrument, which captures the image of scattered pump-laser radiations from free electrons that exist in x-ray channel during the interaction of pump-laser with target-clusters. The TS experimental setup is designed for the purpose of collecting the details of electrons distribution in the x-ray channel. However, fortunately, the TS data provides
bonus experimental details such as the dynamical gas-flow pattern of the DGSN, and pump-laser propagation details during the generation of ultrabright x-ray source. TS image data is presented below in figure 3.3.
Figure 3.3: (a) X-ray source raw-image captured by the TS optical-setup. (b) Shows that the source edge-points are extracted. (c) The extracted source edge-points and corresponding row vectors are fitted to a linear straight-line. The red-arrows show the high intensity focal-plane. (d) The x-ray image captured by the 50 µm aperture x-ray camera.
The TS image-data clearly shows that the converging pump-laser beam towards the focal-plane and the diverging beam from the focal-plane. The outline points of converging and diverging beams (shown in yellow-color in the above figure 3.3 (b)) are extracted from the TS image data and plotted against the pixel-numbers (or raw-vectors). The plotted graphs are fitted to straight-lines, as shown above in the figure 3.3 (c) and extrapolated to intersect each other. The two intersection points of the extrapolated lines should fall, theoretically, on the focal-plane of the focal-spot. The high-intensity focal-spot waist-size is estimated, as shown in the above figure 3.3 (c), by measuring the distance between two intersection points. The estimated focal-spot waist size is approximately 10 µm. Converging angle is 6.5 and diverging angle is 9 degrees. The Rayleigh range is estimated from the divergence angle (9 degrees) and the definition of Rayleigh range. Estimated Rayleigh range of the high-intense focal-spot is 45 µm.

Presence of high-intensity ultrabright x-rays will alter free electrons distributions in the x-ray channel. The estimated converging / diverging angles of the focusing / defocusing beam from the high-intensity x-ray channel TS picture will be different from the true values of the converging / diverging angles of the TS image for high-intensity focal-spot waist-size estimation. Therefore, estimating focused-beam-waist from the TS image, which has presence of high-intensity x-ray radiation might deceive the experimentalist and will be a meaningless measurement. In order to remove the nonlinearity effects due to the x-ray radiation, the experimental conditions are carefully tuned in such a way to minimize presence of the x-ray radiations in the TS channel. Such TS image is presented in the above figure 3.3 (d), along with the triplet-aperture x-ray
camera images, which clearly shows the reduction of x-ray radiation in the particular TS channel.

The **pulse-width** inside experimental chamber is estimated using a TG FROG device and the average **pulse-width** is 350 fs. Using all above estimations all possible pump-laser parameters at the focal-plane during the real experimental situations are estimated and categorically presented below in the table 3.1.

<table>
<thead>
<tr>
<th>Pump-laser parameters at focal-plane</th>
<th>Pump-laser parameters value at focal-plane</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength ($\lambda_L$)</td>
<td>248.6 nm (4.99 eV)</td>
</tr>
<tr>
<td>Average energy (En)</td>
<td>~ 250 mJ</td>
</tr>
<tr>
<td><strong>Pulse-width</strong></td>
<td>~ 350 fs</td>
</tr>
<tr>
<td>Average power ($P_L$)</td>
<td>~ 1 Terra watts ($10^{12}$)</td>
</tr>
<tr>
<td>Focal-spot waist-size</td>
<td>10 $\mu$m</td>
</tr>
<tr>
<td>Rayleigh range</td>
<td>45 $\mu$m</td>
</tr>
<tr>
<td>Average intensity ($I_L$)</td>
<td>$9 \times 10^{17}$ W/cm$^2$</td>
</tr>
<tr>
<td>Average number of photons</td>
<td>$31 \times 10^{16}$ at 250 mJ</td>
</tr>
<tr>
<td>Electric filed strength ($E_L$)</td>
<td>$7.5 \times 10^7$ V/cm at $I_L$</td>
</tr>
<tr>
<td>Magnetic filed strength ($B_L$)</td>
<td>2500 T at $I_L$</td>
</tr>
</tbody>
</table>

Table 3.1: Pump-laser parameters at the focal-plane.

### 3.3 Theoretical Estimations to Determine Target Parameters

Understanding basic phenomena involved in laser-matter interactions relies on the knowledge about the target, which generates ultrabright x-rays. In XRIM Xe and Kr clusters are the lasing-medium. The clusters are produced by adiabatic expansion of target-gas inside vacuum through a sonic solenoid-nozzle. The experimental measurements of cluster-density and cluster-size produced by different types of nozzles are presented in reference 41 along with the nozzle’s details. The nozzle used for the experiments presented in this thesis is an upgraded version of previously used sonic-nozzles. The nozzle is termed, in this thesis, as Double Gas Sonic Nozzle (DGSN). Any
types of experiments were not performed to characterize density and cluster-size produced by the DGSN. Therefore, in this thesis, simple theoretical estimations are performed to determine (i) number of gas-molecules emitted by the DGSN during the single-shot solenoid poppet actuation (ii) cluster-size produced by the DGSN.

3.3.1 Estimating Number of Gas Molecules Sprayed by DGSN

In this section, an experimental method is presented to estimate number of target-gas molecules emitted by the DGSN during its single-shot operation. In this method, after D chamber is pumped down and isolated from vacuum pumps, the DGSN is operated and D chamber vacuum level is recorded for every 10 shots.

The ideal gas equations before the operation of DGSN is given by

\[ P_{\text{initial}} V_{\text{Cha}} = n_{\text{initial}} R T_g \]  

(3.19)

Where \( P_{\text{initial}} \) is D chamber initial pressure, \( V_{\text{Cha}} \) is volume of D chamber, \( n_{\text{initial}} \) is initial number of gas molecules inside D chamber (it is assumed only target-gas molecules are inside chamber), \( R \) is the ideal gas constant (8.314 JK\(^{-1}\)mol\(^{-1}\)), \( T_g \) is the target-gas temperature (293 K).

After the DGSN is operated \( No \) times, the ideal gas formula for the D chamber could be rewritten as

\[ P_{No} V_{\text{Cha}} = n_{No} R T_g = (n_{\text{initial}} + No \times n_g) R T_g \]  

(3.20)

Where \( P_{No} \) is the D chamber pressure after the nozzle is operated \( No \) times, \( n_g \) is number of gas molecules emitted by the DGSN during its single-shot operation and all other parameters has previously defined meanings.
Dividing equation 3.20 by the equation 3.19 and mathematical manipulation leads to an equation presented below in equation 3.21

$$\Rightarrow P_{No} = P_{initial} - \left( \frac{P_{initial} \times n_g}{n_{initial}} \right) No$$

(3.21)

As shown below in figure 3.4 values of $P_{No}$ is plotted against $No$ and fitted to a linear-straight-line.

From the equation 3.21, the slope (Slope) of the graph could be mathematically expressed by a expressions presented below.

$$\text{Slope} = \left( \frac{P_{initial} \times n_g}{n_{initial}} \right)$$
This leads to
\[
\Rightarrow n_g = \left( \frac{\text{Slope} \times n_{\text{initial}}}{P_{\text{initial}}} \right)
\]  
(3.22)

Combining equation 3.22 and equation 3.19
\[
n_g = \left( \frac{\text{Slope} \times V_{\text{Cha}}}{R T_g} \right)
\]  
(3.23)

In the equation 3.23 Slope (0.35 mtorr) is estimated from the graph presented in the above figure 3.4 and volume of the D chamber \(V_{\text{Cha}}\) is 0.51 m\(^3\). \(R\) is the ideal gas constant with value \(8.314 \times 10^{-5} \text{ m}^3\text{barK}^{-1}\text{mol}^{-1}\). Substituting these values in the equation 3.23 will lead to estimate number of gas molecules \(n_g\) emitted by the DGSN during its single-shot operation. The estimated value for \(n_g\) is \(30 \times 10^{17}\) target-gas molecules.

The gas-molecules emitted from DGSN forms clusters and increases density of target-material for laser-matter interaction. The following section estimates cluster-size produce by the DGSN.

### 3.3.2 Estimating Target Gases Clusters Size and Density

Clusters produced from rare-gas cooling in sonic expansion constitute an intermediate state of matter that combines advantages of gas-targets with an absorption efficiency that is even greater than that of solid-targets [50]. In 1956, Becker, Bier, and Henkes gave first report on the formation of clusters by condensation of the respective vapor expanding out of a miniature nozzle into vacuum [51]. Today clusters play important role in various fields. Since formation of clusters was discovered, there have been so many experimental and theoretical approaches proposed to characterize the
clusters produced by sonic-nozzles. The following section discusses clusters formation and theoretical estimations of clusters-size and structures.

3.3.2.1 Clusters Size and Structure

When a high-pressure gas flows into vacuum through an orifice, its expansion is isentropic. The random thermal-energy is converted into the directed kinetic energy resulting in a decrease in temperature according to the Joule-Thompson effect. For certain gases, the gas can become supersaturated, and liquid or solid density droplets will form. These droplets are bonded by Van der Waals forces [52]. Clusters occupy the region between atoms/molecules-monomers-and condense phase. Thus, clusters-size range from the dimmer up to microcrystal or micro-droplets [51]. Cluster-size (N) plays important role during the laser-matter interaction mechanism. The following example explains how the available interaction-area increased during the clusters formation. A cube with \( N_s = 10^6 \) atoms has only 6 % surface-atoms. In other hand, during the clusters-formation for only 1000 \( (N_s = 1000) \) atoms this surface-fraction increases to 49 %. This clearly shows that even for small number of molecules \( N_s \) almost all atoms are surface-atoms. The surface-atoms are less tightly bound compared to the volume-atoms [51] and the numbers of available target-gas surface-atoms for laser-matter interactions is proportional to the x-ray intensity produced from super-radians mechanism. Therefore, the size of the clusters produced by the sonic-nozzle plays important roles in the experiments for generation of ultrabright x-rays. As mentioned earlier a DGSN produces target-clusters and characterization experiments were not performed for this nozzle to determine clusters-density and size. Theoretical calculations performed to estimate the clusters-size and density is presented below.
There are no complete theories available for estimating exactly the clusters-size and density of clusters produced by nozzles. An empirical formula was proposed by Hagena in order to estimate the average clusters-size [51] and [52]. The formula first established from mass spectrometer measurements [51] and then confirmed via different techniques including high-energy electron diffraction, He atom scattering, and Rayleigh scattering on argon (Ar) clusters [50]. In this thesis, size of clusters produced by the DGSN is estimated from the Hagena empirical formula. As a first step, the dimensionless Hagena parameter $\Gamma^*$ is estimated [52] using the equation 3.24 presented below.

$$\Gamma^* = k \frac{P_0}{T_0} \left( \frac{d_t}{\tan \alpha} \right)^{0.85}$$  \hspace{1cm} (3.24)

Where $k$ is the empirical parameter termed as condensation parameter depends on type of gases used (for Xe $k = 5500$ and for Kr $k = 2890$ [52]), $P_0$ is the backing-pressure (in bar), $T_0$ is the gas temperature (in K), $d_t$ is the throat-diameter (in $\mu$m), and $\alpha$ is the half-angle as shown below in diagram 3.1.

Diagram 3.1: Sonic-nozzle and gas flow parameters.

The typical value for $P_0$ is 10.34 bar (150 psig), $T_0$ is 296 K, and $d_t$ is 2500 $\mu$m. The parameter $\alpha$, the half-angle, is estimated from the interferogram presented in the figure 2.3.2 of the chapter 2. Maximum value for parameter $\alpha$ is ~ 10 degrees.

Secondly, the average clusters-size $N_{ci}$ is estimated [53] using the scaling law presented below in equation 3.25.
The Xe cluster-size produced by the sonic-nozzle is 333 atoms at $T_0 = 290$ K and $P_0 = 160$ psig. The Kr cluster-size produced by the sonic-nozzle is 105 atoms at $T_0 = 290$ K and $P_0 = 160$ psig. The target-cluster-size is estimated for different experimental target-gas temperatures. The variation of Xe clusters-size with Xe gas-temperature and the variation of Kr clusters-size with Kr gas-temperature are presented in a single graphical format in the figure 3.5 presented below.

![Graph](image.png)

Figure 3.5: Xe, Kr clusters sizes for different target-gas temperatures.

Determining exact shapes of target-gas clusters formed by the sonic-nozzle is very complicated physics problem. In this thesis, it is assumed that the shape of the
target-clusters formed by the DGSN is sphere. In the cluster-size estimations presented above, the average number of Xe molecules in a cluster at room temperature (293 K) is 296 and Kr molecules in a cluster at room temperature (293 K) is 94. This reasonable number of target-molecules packed together is convincing that the above assumption (Xe/Kr cluster shape is a sphere) is a reasonable assumption. An equation is borrowed from reference 54 and presented below in equation 3.26 to estimate the average clusters diameter ($D_{clstr}$). This is a typical expression used in solid-state / condense-matter physics to express how the atoms are packed in solid states.

$$D_{clstr} \approx 2r_{mole} \times \sqrt[3]{(f_i N_{cl})}$$

(3.26)

Where $f_i$ is the packing-factor of the atoms in the cluster and $r_{mole}$ is the radius of a one target-atom. The radius of a single Xe atom is 108 pm (1 pm = 10^{-12} m) [54] and the radius of a single Kr atom is 88 pm. The packing-factor is the ratio between the volumes occupied by the atoms in a unit-cell to the volume of that unit-cell. The calculated value for packing-factor ($f_i$) of Xe cluster presented in reference 54 is borrowed to calculate Xe cluster-radius. The packing-factor ($f_i$) for Xe cluster is 0.2. It is assumed that that packing factor ($f_i$) for Kr clusters is also same (0.2) as both Xe and Kr are packed in similar fashion. The estimated diameter of a Xe cluster ($D_{clstr}^Xe$) from the equation 3.26 is ~ 1 nm and the estimated diameter ($D_{clstr}^Kr$) of a Kr cluster from the equation 326 is ~ 0.6 nm.

The estimated focal-spot waist-size is presented in the table 3.1 and it is ~ 10 µm. The focal-spot waist-size divided by the cluster-radius will result in a number of clusters packed inside the cross-section of focal-spot waist. Multiplying the average number of
atoms in a cluster with the number of clusters packed inside the cross-section of focal-spot waist-size will result to estimated number of target-molecules packed inside the cross-section of focal-spot waist. Numbers of pump-laser photons that fall on the focal-plane is previously estimated and it is $\sim 31 \times 10^{16}$ at 250 mJ averaged pump-laser energy. Number of pump-laser photons available for exciting each and every atom packed inside the first-Rayleigh-range plane is estimated and presented in table 3.2 below. Similarly, the volume trapped inside Rayleigh-range was estimated and the estimated value is about $10^4 \mu m^3$. Number of target-clusters packed inside the Rayleigh-range volume is estimated and presented in table 3.2. The atoms-packed inside Rayleigh-range volume interact with the pump-laser and will contribute for generation of ultrabright x-rays. Subtracting number of target-atoms packed inside Rayleigh-range volume from number of atoms sprayed by DGSN will result to estimate number of unionized target-atoms. The unionized target-atoms will attenuate propagating x-rays. The ratio of number of target-atoms inside Rayleigh-range to out-side Rayleigh-range is also presented below in table 3.2.

<table>
<thead>
<tr>
<th></th>
<th>Xe</th>
<th>Kr</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of atoms in a cluster</td>
<td>296</td>
<td>94</td>
</tr>
<tr>
<td>Cluster-diameter ($D_{clstr}$)</td>
<td>1 nm</td>
<td>0.6 nm</td>
</tr>
<tr>
<td>Number of target-atoms in the focal-spot waist</td>
<td>$10^4$</td>
<td>$1.6 \times 10^4$</td>
</tr>
<tr>
<td>Number of target-atoms inside the Rayleigh range volume</td>
<td>$26 \times 10^{12}$</td>
<td>$121 \times 10^{12}$</td>
</tr>
<tr>
<td>Number of target gas molecules sprayed by DGSN during its single-shot operation</td>
<td>$30 \times 10^{17}$</td>
<td>$30 \times 10^{17}$</td>
</tr>
<tr>
<td>Ratio of No atoms sprayed by DGSN to No. of atoms trapped inside-side Rayleigh-range volume.</td>
<td>$1:1 \times 10^5$</td>
<td>$1:3 \times 10^4$</td>
</tr>
<tr>
<td>Number of pump-photons available for ionizing each target-atom those are inside Rayleigh-range at 250 mJ.</td>
<td>$1 \times 10^5$ ($\sim 5$ MeV)</td>
<td>$3 \times 10^4$ ($\sim 150$ KeV)</td>
</tr>
<tr>
<td>Pump-power per atom those are in Rayleigh range volume</td>
<td>$\sim 1$ W/atom</td>
<td>$\sim 1$ W/atom</td>
</tr>
</tbody>
</table>

Table 3.2: Target-material parameters inside Rayleigh-range.
The interaction of laser with matter is a very complicated physical mechanism. The above estimations ease the understanding the mechanisms involved during the interaction of pump-laser with matter in XRIM experiments. The following section 3.4 discusses the fundamental physical processes involved during the interaction of pump-laser with matter.

### 3.4 Generation of Ultrabright X-rays from Laser Matter Interactions

Understanding the physics behind the laser-matter interactions phenomena will lead to optimize the x-ray source. This section 3.4, generation of ultrabright x-ray from laser-matter interactions, presents theoretical models and theories to understand the interaction of laser with matter in XRIM experiments. As stated earlier, this section 3.4 also adopts previously developed theories and models of XRIM to explain the experimental observations of the experiments performed in the D chamber. Understanding the physics behind the laser-matter interaction demands details of target-material as well as details of pump-laser. The following section 3.4.1 discusses details of target-material.

#### 3.4.1 Details of Target Material

Selection of suitable target-material is the key factor for the success of generation of ultrabright x-ray sources. Theories and experiments predict that the gases with higher-atomic numbers are suitable material for generation of ultrabright x-rays. Radon (Rn) is the densest gas among the noble gases at room temperature with atomic number 86 and molecular or atomic weight of 222. Accordance with periodic trends, Radon has a lower
electronegativity than the element Xe one before it and it is more reactive and radioactive gas. Radioactive and reactive natures of Rn gas prevent using it as target-material in laboratories. In the periodic table, the two noble gases just before Rn are Xe and Kr. They are dense enough, non-reactive, and non-radioactive stable gases at room temperature.

Further, the elements with even atomic number are suitable for using it as lasing medium. The reasons are presented as follows. The elements with odd-atomic number have nuclear spin and nuclear moment and those with even number tend to have no nuclear spin, one possible explanation for this anomalous behavior is that hyperfine splitting plays an important role in the gain of (neon-like laser) lines for elements with odd-atomic number [55] and [56]. The hyperfine splitting can affect the gain of the laser-line by effectively increasing the line-width. Since the gain is inversely proportional to the line-width, the gain will decrease if the width increases. If the splitting is large enough, a single line may be split into several weaker lines. Therefore, even atomic number gases such as Xe and Kr are used as target-material for generation of ultrabright x-rays. The target-material understanding is the very first step to understand the mechanisms involved in the interactions of laser with matter. A brief introduction to Xe and Kr target-materials are presented below separately in the following sections.

3.4.1.1 Xenon

Xenon was discovered in England by the Scottish chemist William Ramsay and English chemist Morris Travers on July 12, 1898, shortly after their discovery of the elements Krypton and Neon. They found Xenon in the residue left over from evaporating components of liquid air. Ramsay suggested the name xenon for this gas from the Greek
word ξένον, neuter singular form of ξένος (Xenos), meaning foreign(er), strange(r), or guest.

Atomic-number of Xe is 54 and mass-number is 131. The electronic configuration of the Xe atoms is 1s² 2s² 2p⁶ 3s² 3p⁶ 4s² 3d¹⁰ 4p⁶ 5s² 4d¹⁰ 5p⁶. The ionization potential of Xe is presented below in the appendix 3.1 and a tentative Xe \textit{M-shell} and Xe \textit{L-shell} wavelength range are also presented at the end of the appendix 3.1. The absorption edges of the Xe atoms are presented in the appendix 3.2.

3.4.1.2 Krypton

Two scientists, Sir William Ramsay, a Scottish chemist, and Morris M. Travers, an English chemist discovered Krypton while studying liquefied air on May 30, 1898 just few months before they discovered Xe.

Atomic mass of Kr is 36 and mass-number is 84. The electronic configuration of the Xe atoms is 1s² 2s² 2p⁶ 3s² 3p⁶ 4s² 3d¹⁰ 4p⁶. The absorption edges of the Kr atoms are presented in appendix 3.3.

The following section 3.4.2 briefly discusses the ionization mechanism that takes place during the interactions of laser with matter.

3.4.2 Ionization Process

The ionization process begins with the interactions of laser light with target medium. The very first efforts to understand the interactions of light with matter were performed by Alexandre Edmond Becquerel during the discovery of photovoltaic effect on electrolytic cells in 1839. The puzzles of interactions of light with matter were remained unsolved even after the great scientist James Clerk Maxwell formed four
Maxwell equations in the early 1860’s to describe electromagnetic waves. In 1887, Heinrich Hertz observed the photoelectric effect, the emission of electrons from matter as a consequence of their absorption of energy from electromagnetic radiation of very short wavelength, such as visible or ultraviolet radiation. Albert Einstein proposed revolutionary idea of light quanta and mathematically explained the photoelectric effect in 1905. Theory of photoelectric is one of the first theoretical works explained interaction of light with matter. In 1914, Robert Andrews Millikan experimentally confirmed the Albert Einstein theory of photoelectric effect.

The discovery of lasers increased tremendously the available light-intensity and challenged the validity of the theory of photoelectric at higher-intensities where the electric-filed of light is higher than the coulomb electric-field strength of atoms. The high intensity laser-matter interactions experiments performed after discovery of lasers showed that the emission of electrons from target-material is possible even when the photon energy is lower than the work function of that target-material. Albert Einstein’s theory of photoelectric effects fails to explain this phenomenon. The experimental observation is explained later by the phenomena of multiphoton photoemission and modification of effective work function of target-material. Further, the laser-matter interactions experiments revealed that new physical phenomena such as nonlinear photoionization, multiphoton ionization, tunneling ionization taking place during the interactions of high intensity-laser with matter and the theory of photoelectric effect were not enough to explain these phenomena. Therefore, completely new theories are necessary to understand the physical phenomena of high intensity laser-matter interactions and strong-field atomic physics.
In strong-filed atomic physics such as the high-intensity laser interaction with matter the strength of field is sufficiently strong enough to compete with the coulomb binding-filed in controlling the atomic electron dynamics. A simple estimation presented below elaborates it clearly.

The relationship between laser intensity \( I_L \) and electric filed \( E_L \) strength could be given by equation 3.27 [57].

\[
I_L = \frac{1}{2} \varepsilon_0 c E_L^2
\]  

(3.27)

Where \( \varepsilon_0 \) is the permittivity of the free space and \( c \) the speed of light. Simplified electric-filed strength expression is presented below in equation 3.28.

\[
E_L = 27 A \sqrt{I_L}
\]  

(3.28)

The coulomb electric field in a hydrogen atom is \( E = e/\left(4\pi\varepsilon_0 a_0^2\right) \approx 5 \times 10^9 \text{ V/cm} \) (where \( a_0 \) is the radius of hydrogen atom), which corresponds to an equivalent laser-intensity of \( 3.51 \times 10^{16} \text{ W/cm}^2 \). The laser-matter interaction in high-field strength depends on the kinetic-energy of the electrons oscillating in the field, and the ionization energy of the atoms / ions. At the laser electric-filed strength, \( 5 \times 10^9 \text{ V/cm} \) the magnetic-field strength is about \( 10^3 \text{ Tesla} \) and the velocity of electrons could reach to 1/100 times speed of light. Therefore, at the laser intensity regimes where the laser electric-filed is higher than the atomic field-strength the atoms are sufficiently perturbed and interesting nonlinear photoionization phenomena takes place.

Since the discovery of ruby laser, pulse amplification technologies of laboratory-scale lasers faced at least four development phases; free running, Q-switching, mode locking and chirped pulse amplifications. Every time the pulse amplification technologies
boosted laser-intensity-level, the strong field effect also opened a new window to observe new multiphoton phenomena. In 1960, a group at Saclay, France led by Manus and Mainfray [57] observed the first multiphoton ionization. The second development took place in the late 1970’s and 1980’s. In 1979, the third development, Above Threshold Ionization (ATI) observed. The fourth and recent phenomenon is the tunneling ionization. All four phenomena are briefly described below.

### 3.4.2.1 Multiphoton Ionization Mechanism

The Multiphoton ionization (MPI) is the multiphoton transition from a bound-state to a free (continuum)-state. If the ionization potential of atoms is greater than the photon energy, the atoms have to absorb several photons to be ionized, and hence this process is called MPI. The energy level structure for MPI is presented below in diagram 3.2 (a).

![Diagram 3.2: (a) Energy-level diagram for MPI. (b) Energy-level diagram for ATI.](image_url)
3.4.2.2 Above-Threshold Ionization Mechanism

An emitted free electron in a laser-field possesses, in addition to any translational kinetic energy, quiver energy due to the oscillatory motion imparted on it by the laser-field. The average kinetic energy associated with the quivering force upon an electron by a radiation field acts as a potential for the average motion of the electron. The quiver energy is known as ponderomotive energy $U_p$, and given below by equation 3.29.

$$U_p = \frac{e^2 E^2_L}{4m_e \omega_L^2}$$  \hspace{1cm} (3.29)

Where $e$ is the charge of the electron, $E_L$ is the local instantaneous electric-field, $m_e$ is the mass of the electron and the $\omega_L$ is angular frequency of the laser-field.

In general, ponderomotive potential $U_p$ is time dependent. Therefore, the total kinetic energy is not a constant of the motion, and the ponderomotive potential $U_p$, in general, is also not a conserved parameter. In terms of laser wavelength $\lambda_L$ (in nanometers) and laser-intensity $I_L$ (in W/cm$^2$) the ponderomotive energy $U_p$ (in eV) equation could be rewritten as presented below in equation 3.30.

$$U_p = (9.33 \times 10^{-20}) I_L \lambda_L^2$$  \hspace{1cm} (3.30)

The ponderomotive energy produces noticeable effect on bound electrons when the laser field is intense enough. Since Rydberg levels are weakly bound at very intense-fields, their induced shifts are very close to the ponderomotive energy $U_p$, while states nearer the nucleus have much smaller polarizability and will be harder to influence. Therefore, they have, correspondingly, smaller shift. Typically, the Rydberg series and continua shift to such an extent that the shift of the very low states can be neglected. This
means that the ionization potential increases by approximately $U_p$. This increase, if large enough, will prohibit ionization through the low-order channels as presented in the above diagram 3.2(b) [57]. Therefore, when an intense laser pulse ionizes an atom or molecule, it needs more photons than necessary to overcome the ionization threshold. This phenomenon is called Above Threshold Ionization (ATI).

The ATI phenomenon is intimately related to the non-sequential ionization and high order harmonic generation. The first series of experiments on the excitation of clusters by a femtosecond laser-pulses for understanding the ATI phenomenon were carried out by the McPherson et al in XRIM laboratory in 1987 [57].

The value for ponderomotive potential $U_p$ is calculated using the equation 3.30 by substituting the pump-laser parameters. The calculated ponderomotive potential $U_p$ is $6 \times 10^{-3}$ eV.

3.4.2.3 Tunneling Ionization Mechanism

In 1965, Lenoid Veniaminovich Keldysh proposed that tunneling ionization could occur under certain conditions. At modest laser intensities, the normal multiphoton excitation route for ionization via intermediate states applies. If the incident field is strong enough and frequency low enough, then within a quasi-stationary approximation, the field is able to distort the atomic-potential to such an extent that a potential barrier is formed through which the electron can tunnel. This phenomenon is known as tunneling ionization. If the field-strength is increased further the interaction term becomes stronger and so makes the gradient increasingly more negative. This means that the barrier becomes smaller and lower until eventually the ground state is no longer bound. This is
known as Over The-Barrier Ionization (OTBI) [57]. All of the above-discussed three scenarios are presented below in figure 3.6.

\[ I < 10^{14} \text{ W cm}^{-2} \]

Figure 3.6(a) \[ I < 10^{15} \text{ W cm}^{-2} \]

Figure 3.6(b) \[ I > 10^{15} \text{ W cm}^{-2} \]

Figure 3.6(c)

Figure 3.6: Three types of ionization mechanisms (a) Multiphoton, (b) Tunneling, (c) Over the barrier.

In the OTBI (in W/cm²), the critical field is obtained by equating the maximum induced by the field in the atomic potential to the binding-energy [57]. The critical filed for OTBI leads to critical intensity given in equation 3.31 presented below.

\[
I_{\text{OTBI}} = \frac{\pi^2 e^3 I_p^4}{2Z^2 E_L^6} = \frac{4 \times 10^9 (I_p)^4}{Z^2} \quad (3.31)
\]

Where \( Z \) is the charge-state of the target-atom or ion and \( I_p \) (in eV) is the ionization potential of the target at the charge-state \( Z \) and all other parameters have the same meaning as previously defined.

The **pulse-width** of pump-laser could be defined as the amount of time between the pulse’s maximum intensity and the time the intensity drops to 1/e of the maximum value [58]. During the interaction of laser with matter, the pump-laser pulses begin to interact with matter in the leading edge of the pulse. After the laser-pulse reaches the maximum intensity, it drops to zero at the tailing edge of the pulse. If pump-laser **pulse-width** is a longer pulse and the peak intensity is much higher than the intensity for
tunneling ionization, the integrated effect of MPI at the raising part of the pulse would be enough to almost (or totally) ionize the atom. Thus, before the pulse’s intensity reaches the regime of tunneling ionization, there are no more neutral atoms left to interact with the high-intensity part of the pulse. Further, the tunneling ionization mechanism does not take place in the very beginning of the laser pulse due to small values of the laser field strength $E_L$. Therefore, the tunneling ionization or OTBI requires ultrashort laser pulses to preempt the preionization.

The critical intensity values for the OTBI are calculated for the Xe M-shell and L-shell charge-states using the equation 3.31 and presented in a tabular format in appendix 3.4. Estimated critical intensity values for the Xe M-shell lies in the range between $10^{19} - 10^{20}$ W/cm$^2$ and L-shell $10^{21} - 10^{24}$ W/cm$^2$. It is clear that the pump-laser intensity ($I_L = 10^{17}$ W/cm$^2$) produced in the XRIM is not enough to excite L-shell electrons including 2p electrons through tunneling ionization or OTBI.

In 1965, Keldysh defined a parameter termed Keldysh parameter $\gamma$, which determines whether multiphoton or tunneling ionization dominates the nonlinear ionization process. The Keldysh parameter $\gamma$ is the ratio between the frequency of the laser light and the frequency of the electron tunneling through a potential barrier. The tunneling ionization of atomic states dominates when the Keldysh parameter $\gamma \ll 1$ ($I_p < U_p$), while for the Keldysh parameter $\gamma \gg 1$ ($I_p > U_p >> \hbar \omega$) most significant ionization is multiphoton process. An expression for the Keldysh parameter $\gamma$ is presented below in equation 3.32 followed by the calculation to determine the Keldysh parameter $\gamma$ for the pump-laser of XRIM.
\[ \gamma^2 = \frac{\omega_L^2}{\omega_{\text{tunneling}}^2} = \frac{e^2}{e^2 \times E_L^2} \times 2m_e I_p = \frac{I_p}{2U_p} \]  

(3.32)

Where \( \omega_L \) is the frequency of laser-light, \( \omega_{\text{tunneling}} \) is frequency of electron tunneling though potential barrier, \( I_p \) is ionization potential, \( m_e \) is mass of electron, \( e \) is charge of electron, \( E_L \) is the field strength of the pump-laser, and \( U_p \) denotes the mean energy transferred to an electron in its oscillatory motion caused by the electromagnetic laser field.

The Keldysh parameter \( \gamma \) is calculated using the equation 3.32 for different charge-states \( Z \) and presented in the appendix 3.4. The presented \( \gamma \) for different charge-states implies that tunneling ionization is not a possible ionization mechanism for the laser parameters presented in the table 3.1.

An expression for the tunneling ionization probability \( W \) of the Xe\(^{26+} \) charge-states, using ADK (Ammosov, Delone, and Krainov) approach, is presented below in equation 3.33 [52].

\[ W = \Delta t \left( \frac{e}{\pi} \right)^{3/2} \sqrt{3} \left( \frac{2I_p}{Z} \right)^{3/4} \left( \frac{16eI_p^2}{ZE_L} \right)^{2/3} \sqrt{i} \text{exp} \left[ - \frac{2(2I_p)^{3/2}}{3E_L} \right] \]  

(3.33)

Where \( \Delta t \) is the temporal duration of the action for the given field strength value \( E_L \), and \( I_p \) is the ionization potential of an atomic ion with the charge multiplicity \( (Z - 1) \).

Calculated tunneling ionization probability \( (W) \) for the Xe\(^{26+} \) gives very small value (~ 10^-8), which reconfirms that the tunneling ionization does not result in producing Xe atomic ions with \( Z > 26 \).
3.4.2.4 Collisional Ionization Mechanism

Experiments performed in the small-chamber showed ultrabright $L$-shell x-ray radiations from Xe clusters. A potentially very effective method of producing an extremely bright source of $L$-shell x-ray radiation would be to generate a controlled population inversion in the 2p inner-shells of high-Z Xe atoms. Generation of 2p vacancies for $L$-shell x-ray radiations through field ionizations requires a laser irradiance of $\sim 10^{21} \text{ W/cm}^2$, which is four orders of magnitude greater than expected focal-intensities. Therefore, there should be a different mechanism, which is responsible for ionizing Xe $L$-shell. There are various theoretical ideas proposed to explain the experimental observations. One of the mechanisms is based on the selective collisional ionization of inner-shell electrons by ponderomotively-driven electrons initially photoionized by strong above threshold ionization (ATI). The selectivity relies on the fact that the wave function of a photoionized electron distribution maintains its phase and geometric symmetry for a time shorter than the characteristic collisional dephasing time that is in the order of femtosecond[24]. The collisional ionization mechanism is briefly discussed below.

The collisional ionization mechanism is explained in the reference 24 using a relativistic classical model. In the model, it is shown that under influence of the 248.6 nm irradiation with the intensity range $10^{18}$-$10^{19}$ W/cm$^2$ the ponderomotively-driven electrons (ionized from more weakly bound-orbital ionized by ATI) can return to their cluster of origin with an energy of 10-20 k eV; a value sufficient to eject 2p electrons collisionally and thus produce the $L$-shell x-ray emissions. The efficiency of the inner-
orbits ionizations due to collisional ionization is not only decided by the energy of the incident electrons but also determined by the electrons scattering cross-sections.

A mathematical expression for the scattering cross-section for an electron to scatter with an inner-shell electron was derived by Bethe in 1930 [59]. The Bethe scattering expression is presented below in equation 3.34.

\[
\sigma_{nl} = \frac{2\pi e^4}{m_e v^2 E_{nl}} Z_{nl}b_{nl} \ln \left( \frac{2m_e v^2}{B_{nl}} \right)
\]

(3.34)

Where \( E_0 = \frac{1}{2} m_e v^2 \) is the energy of the incident electron, \( E_{nl} \) is the binding energy of the electrons in the \( nl \) shell, \( Z_{nl} \) is the number of electrons in that shell. The parameter \( b_{nl} \) was estimated by Bethe using hydrogenic wave function to be between 0.2 and 0.6 for inner-shells and, for a given shell, to be a function of \( Z \). The energy \( B_{nl} \) was estimated by Bethe to be of the order of \( E_{nl} \).

Using the modified Bethe scattering expression and scattering cross-sections research works presented in reference 60 the magnitude of the scattering cross-sections evaluated from a single plane-wave analysis for the ejections of inner-shell electrons from 2p, 3s, 3p, and 3d orbits. Further, details are presented in the reference 24 and the graph of scattering cross-sections for the ejections of 2p, 3s, 3p, and 3d electron as the function of the incident colliding energy is presented below in figure 3.7.
The figure 3.7 shows that the collisional ionization mechanism is capable of ionizing \textbf{L-shell} of Xe atoms. The figure 3.7 further indicates that an electron incident on a Xe ion with a collisional energy of 10-20 keV is \sim 10 times more likely to eject a 3d, 3p, or 3s electron than a 2p electron with ionization potential nearly equal to 8 keV. If the electron is incident with energy equal to \(I_p\), it must undergo an exactly head-on collision with the inner-core electron for both electrons to escape from the atom or ion. The low probability of such a collision is dramatically increased if the incident electron has more energy since the conditions for collisional ejections are relaxed. However, for collisional energy greater than 10\(I_p\), the probability of inner-core ejection is again diminished due to reduced collision time. Therefore, according to the first Born approximation of plane-wave collisional analysis the experimental observation of ejection of 2p electron before ejection of 3d electrons is an impossible ionization mechanism as discussed below. The equation 3.33 is the fundamental equation for the plane-wave analysis and it is valid.
when $E_0$ is much greater than $E_{nl}$ and when the energy transfer in the ionizing collisions is much less than $E_0$. The ionization energy $E_{nl}$ of the L-orbits is relatively high and comparable to the electrons incident energy $E_0$, which makes the plane-wave collisional ionization analysis a failure to explain the selective collisional ionization mechanism of the L-orbits. Therefore, in order to explain the Xe 2p population inversion produced by selective collisional ionization mechanisms and some other experimental observations of small-chamber, various analysis methods were proposed. In the reference 24, different analyses methods are proposed to explain previous experimental observations including the selective ionization mechanisms observed in the experiments performed in the small-chamber. This thesis does not present all previous analysis works performed in XRIM to explain the experimental data produced in the small-chamber. In other hand, it borrows previously developed theories and models to explain the experimental observation of the experiments performed in the chamber D. One of such theoretical model presented in references 61, 28 is borrowed to explain experimental observation of experiments performed in the D chamber. The borrowed theoretical models is remodeled and presented below.

### 3.4.2.5 Intra-cluster Scattering Model

A model termed as intra-cluster model is developed by XRIM to explain experimental observations of ultrabright x-rays experiments. The abstract of the intra-cluster process is the action of inelastic-scattering process produced by ionized electrons originating from atoms in the cluster that are accelerated by the external optical-field and collide with other atoms or ions in the system and producing inner-shell excitations. The
detail of the model is presented in references 61, 28, and 62. The essence of the intra-cluster model is borrowed from the references to explain the experiment presented in this thesis. The details of the model are presented below.

In the model, the rare gas is considered as a spherical cluster of \( n \) identical atoms with inter-atomic spacing \( r_0 \) and the over all radius of the cluster is \( R \). For \( n \geq 3 \) the radius \( R \) could be described by \( R \approx r_0 n^{1/3} \), which is similar to nucleus analogy.

Consider the laser irradiation (wavelength \( \lambda_L \)) on rare gas cluster-target, which is weakly bounded by Van der Waals forces. If the characteristic dimension \( R \) of a fully ionized cluster is less than the skin-depth \( \delta \), one can assume that the cluster responds to the laser-field as free atoms whose ionization can be estimated with a tunneling picture. The cluster-size and the number of rare gas molecules in a cluster are the limiting factors in the above condition \( R < \delta < \lambda_L \).

The appearance intensities for the inner-shell ionization are predicted quite well by a simple theoretical model presented in reference 63. The model consists of a superposition of the coulomb potential with a static-electric-field as shown below in equation 3.35. As the external-field strength is increased, the height of the coulomb barrier is suppressed and ionization occurs when the amount of suppression is equal to the ionization potential of the atom. In this case, the super imposed potential [63] could be written as presented below in equation 3.35.

\[
V(x) = -\frac{Ze^2}{x} - eE_Lx
\]  
(3.35)

Where all the notations in the equation 3.35 have previously defined meanings.
The position of the barrier maximum \( x_{\text{max}} \) is found by setting \( \frac{\partial V(x)}{\partial x} \) equal to zero. Equating \( V(x_{\text{max}}) \) to the ionization potential \( (I_p) \) yields an expression for the critical electric field strength \( (\varepsilon) \).

Differentiating the equation 3.35 and equating it to zero gives

\[
x_{\text{max}} = \sqrt{\frac{Ze}{\varepsilon}}
\]

Substituting \( x_{\text{max}} \) and solving for critical-field-strength \( (\varepsilon) \) leads to equation 3.36 presented below.

\[
\varepsilon^2 = \frac{I_p^4}{16e^6Z^2}
\]

(3.36)

The equation 3.27 relates the intensity of laser to its electric field. At the critical electric-field strength \( \varepsilon \), the intensity of the laser is known as the appearance intensity \( I_{\text{app}} \). Therefore, the appearance intensity \( I_{\text{app}} \) could be written as.

\[
I_{\text{app}} = \frac{1}{2} \varepsilon_0 c \varepsilon^2
\]

(3.37)

By combining the equations 3.36 and 3.37 and with the setting \( \frac{1}{4\pi\varepsilon_0} = 1 \) the appearance intensity \( I_{\text{app}} \) becomes simplified and it is given below in equation 3.38.

\[
I_{\text{app}} = \frac{cI_p^4}{128\pi^6Z^2}
\]

(3.38)

Constrains to the proposed model is derived as discussed below. The proposed model is expected to become invalid if the cluster becomes sufficiently large and /or the external electromagnetic field becomes sufficiently strong. This limit could arise through the action of inelastic-scattering process produced by ionized electrons originating from atoms in the cluster that are accelerated by the external optical-field and collide with
other atoms or ions in the system [61]. Roughly, such intra-cluster process can be considered as

(i) Excitation

\[ e^- + X^{q+} \rightarrow \sigma_{ei} \rightarrow e^- + (X^{q+})^* \]

(ii) Additional ionization

\[ e^- + X^{q+} \rightarrow \sigma_{ei} \rightarrow e^- + e^- + X^{(q+1)+} \]

An approximate estimate of the number \( N_x \) of such excitations or ionizations can be written as the product of the number of electrons produced in the cluster \((nZ)\), the atomic density in the cluster \((r_0^{-3})\), the cross-section \((\sigma_{ei})\) for the inelastic channel under consideration and the characteristic scale-length of the cluster \((R)\) \((\approx r_0 n^{1/3})\) [61]. Therefore, for an additional ionization, the expression for \( N_x \) could be written as

\[ N_x \approx (nZ)\sigma_{ei} r_0^{-3} r_0 n^{1/3} = n^{4/3} Z \frac{\sigma_{ei}}{r_0^2} \]  

(3.39)

The critical cluster-size \( n_c \) is defined by \( N_x = n = n_c \). Substituting \( N_x = n = n_c \) in the equation 3.39 to get the expression for the critical cluster-size \( n_c \) and it is presented below in equation 3.40.

\[ n_c = \left( \frac{r_0^2}{Z \sigma_{ei}} \right)^3 \]

(3.40)

Therefore, if \( \sigma_{ei} \) corresponds to an inner-shell ionization, with \( n \geq n_c \) an inner-shell vacancy would be produced in every rare-gas target (Xe/Kr) atoms.
During the intra-cluster process, certain conditions must be met in order for the inner-shell excitations mechanism to take place. Specifically, inner-shell excitation can only take place if colliding electrons possess sufficient kinetic energy. To estimate this energy, and the conditions necessary for its transfer, it is assumed that the electrons experience acceleration over the characteristic dimension \( R \) of the cluster by external optical-field \( E_L \) in a time short compared to the period of the oscillating wave. Further, it is assumed that if the electrons leave the vicinity of the cluster, they represent ionization and are lost from the system. This simple procedure yields a characteristic energy \( \varepsilon_e \) given by

\[ \varepsilon_e \approx e E_L R \]  

(3.41)

Since \( R \approx r_0 n^{1/3} \) and \( E_L = \varepsilon \) (critical electric field-strength) the equation 3.41 becomes

\[ \varepsilon_e \approx e r_0 n^{1/3} \]

\[ \Rightarrow \varepsilon = \frac{\varepsilon_e}{e r_0 n^{1/3}} \]  

(3.42)

Reorganize the equation 3.36 results

\[ I_p^4 = e^2 16e^6 Z^2 \]  

(3.43)

The equation 3.38 and the equation 3.43 are combined to express the characteristic minimum intensity \( I_0 \) (or appearance intensity \( I_{app} \)) needed for inner-shell exaction. The combined equation is presented below in equation 3.44

\[ I_0 = \frac{e^2}{8\pi} \]  

(3.44)
By combining the equation 3.42 and equation 3.44, the expression for the characteristic minimum intensity \( I_0 \) can be obtained as presented below in equations.

\[
I_0 = \frac{c e^2}{8 \pi e^2 n_0^2 n^{2/3}} \\
\Rightarrow I_0 = \frac{e^2}{8 \pi \left( \frac{e^2}{\hbar c} \right) \hbar n_0^3 n^{2/3}}
\]

Fine-structure constant \( \alpha \) is the coupling constant characterizing the strength of electromagnetic interaction. The Fine-structure constant \( \alpha \) is equal to \( \left( e^2 / \hbar c \right) \). Therefore, the characteristic minimum intensity \( I_0 \) could be simply expressed by equation 3.45 presented below.

\[
I_0 = \frac{e^2}{8 \pi \alpha n^{2/3} \hbar n_0^2}
\]  \hspace{1cm} (3.45)

Since this simple picture assumes that, the acceleration occurs in a time comparable to or less than a period of the wave, validity of this approximation demands that the cluster neither be too large nor the external field too small. It requires that the amplitude \( x_e \) of the driven excursion of the electron caused by the field be comparable to or larger than the cluster-dimension \( R \). If one could set the limiting situation as \( x_e = R \), assume that the electron is free and put \( x_e = eE/m_e \omega_L^2 \), with \( m_e \) denoting electron mass and \( \omega_L \) the angular frequency of the external optical-field, it is possible to define another limiting intensity \( I_n(\lambda) \) as derived below [61].

Cluster-radius is defined previously as \( R = r_0 n^{1/3} \)
Since $x_e = R = eE/m_e\omega_L^2$

$$R = r_0n^{1/3} = \frac{eE}{m_e\omega_L^2}$$

$$r_0n^{1/3} = \frac{eE_L^2}{4m_e\pi^2c^2}$$

and $\omega = \frac{2\pi}{\lambda}$

$$\Rightarrow E_L = \frac{r_0n^{1/3}4m_e\pi^2c^2}{e\lambda_L^2}$$

Substituting the above expression for $E_L$ in the equation 3.41 ($\varepsilon_e \approx eE_LR$) it becomes

$$\varepsilon_e \approx eE_Lr_0n^{1/3} = \frac{r_0^2n^{2/3}4m_e\pi^2c^2}{\lambda_L^2}$$

Substituting $\varepsilon_e$ in the equation 3.45 leading an expression for the second-limiting intensity $I_n(\lambda_L)$. The limiting expression is presented below.

$$I_n(\lambda_L) = \frac{1}{8\pi cn^{2/3}h\lambda_L^2} \times \left( \frac{r_0^2n^{2/3}4m_e\pi^2c^2}{\lambda_L^2} \right)^2$$

$$\Rightarrow I_n(\lambda_L) = \frac{2\pi^3}{\alpha} \times \left( \frac{r_0}{\lambda_L} \right)^2 \left( \frac{m_e^2c^4}{\hbar} \right) \left( \frac{n^{2/3}}{\lambda_L^2} \right)$$

The Compton wavelength $\lambda_c$ is $\hbar/m_e c$. Therefore, second limiting intensity $I_n(\lambda_L)$ becomes

$$\Rightarrow I_n(\lambda_L) = \frac{2\pi^3}{\alpha} \times \left( \frac{r_0}{\lambda_L} \right)^2 \left( \frac{m_e^2c^4}{\hbar} \right) \left( \frac{n^{2/3}}{\lambda_L^2} \right), \ (n \geq 3) \quad (3.46)$$

In the proposed model, an inner-shell electron with binding energy $\varepsilon_e$ can be ionized, if the intensity exceeds a lower bound given by $I_0$ or $I_n(\lambda)$, which ever is greatest. The collisional excitation mechanism discussed above can lead to the emission
of prompt inner (j-1) inner-shell radiation in the x-ray range. Generally, in addition to
producing a hole in the (j-1) inner-shell, the possibility of prompt j \( \rightarrow \) (j-1) transition
requires retention of at least one electron in the j-shell during the course of irradiation.
This determines an upper bound in the intensity of the irradiation \( I(j)_{\text{max}} \) through equation
3.38, which is in the simple model considered, independent of the cluster-size.

The limitations for the inner-shell excitations predicted by the intra-cluster model are
summarized below.

(i) Intensity \( I > I_0 \)

(ii) Intensity \( I > I_n (\lambda_L) \)

(iii) Intensity \( I < I(j) \)

The above three conditions are implemented in graphical format (presented in
figure 3.9 for Xe and figure 3.11 for Kr) along with cluster-size to express the laser
intensity-limits for producing Xe \textit{M-shell} x-rays, Xe \textit{L-shell} x-rays, and Kr \textit{L-shell} x-
rays.

3.4.2.5.1 Intensity and Cluster Size Constrain for the Xe X-rays Generation

Threshold intensity \( I_{\text{max}} \) for ionization from bound states of Xe \cite{62} is presented
below in figure 3.8.
Three predicted conditions \((I > I_0, I > I_n(\lambda), I < I(j))\) of the intra-cluster model is presented in graphical form in the above figure 3.8. Therefore, the figure 3.8 illustrates the conditions for the intensity and cluster-size for which the intra-cluster model allows prompt Xe \textit{M-shell, L-shell} emissions. For particular cluster-size produced by the sonic-nozzle, the laser intensity should fall inside the allowed shaded-areas to produce prompt inner-shell x-rays. The estimated cluster-size produced by the DGSN is 296 Xe molecules per a cluster and it is shown in figure the 3.11 by the yellow line. The minimum corresponding laser intensity for producing inner-shell x-rays is the intersection
of yellow line with $I_0$. The minimum intensities for the generation of Xe $M$-shell and $L$-shell are also shown in the figure 3.9 and the values are presented below.

The minimum intensity limit for Xe $M$-shell x-ray generation is in the order of $10^{16}$ W/cm$^2$.

The minimum intensity limit for Xe $L$-shell x-ray generation is very close to the order of $10^{18}$ W/cm$^2$.

Performed experiments at the small-chamber confirmed the predictions of the intra-cluster model for Xe and Kr clusters [28], and [62]. Performed analysis for Kr is presented below in following section.

![Figure 3.10: Threshold intensities for ionization from bound-states of Kr [62].](image1)

![Figure 3.11: Intra-cluster allowed zones for Kr $M$ and $L$-shell as a function of intensity and cluster-size.](image2)

### 3.4.2.5.2 Intensity and Cluster size Constrains for Kr X-rays Generation

Threshold intensity $I_{\text{max}}$ for ionization from bound states of Kr is presented above in the figures 3.10 and 3.11.
The estimated Kr cluster-size produced by the DGSN is 94. Therefore, according to the intra-cluster model prediction, the minimum intensity limit for Xe L-shell x-ray generation is in the order of $10^{17}$ W/cm$^2$.

In next chapter 4, the intra-cluster is used as a tool to explain Xe, and Kr emission spectral features of spectra collected by von Hámos single crystal spectrometers. Even though the intra-cluster model successfully explain ultrabright x-ray source experimental observations, it is not a complete theory to explain some Xe emission spectral features those were not observed in Kr spectra. In addition to all of the presented models a completely new mechanism, termed as dynamical orbital collapse mechanism is discussed in this thesis to explain the experimental observations (especially Xe spectra features) presented in this thesis. The theory of dynamical orbital collapse is developed by XRIM to explain previous experimental observations of XRIM ultrabright Xe x-rays generation. The idea of orbital collapse is borrowed to explain the experimental observation of $4f \rightarrow 3d$ transitions in Xe. In addition to this explanation, this theory explains why Xe $4f \rightarrow 3d$ transitions is not observed in Kr experiments.

### 3.4.2.6 Dynamical Orbital Collapse Mechanisms

Dynamical Orbital Collapsed (DOC) one of the prominent mechanisms presented in reference 46, explains experimental observations such as abundance of Xe $4f \rightarrow 3d$ transitions. The referred mechanism is an entirely different mechanism of excitation, which automatically optimizes the electronic configuration of the atom for rapid and efficient x-ray emission in the kilovolt range [46]. The details of the dynamical orbital collapsing mechanisms are presented along with the experimental evidences in the
This thesis does not discuss the details of mechanisms but it borrows the idea to explain the experimental data produced in the D chamber during the generation of ultrabright x-ray source.

The dynamical orbital collapse mechanism is a combination of three individual and independent atomic affects, namely, (i) MultiPhoton Ionization (MPI), (ii) nf-orbital collapse arising from the interplay of shielding and the centrifugal barrier on the intra-atomic potential, (iii) electron shake processes, which causes the redistribution of electrons in the bound states of ions through a direct intra-atomic electron-electron interaction. The MPI is already discussed earlier. The brief details of the nf-orbital collapse and the electrons shake process mechanisms are presented below.

**Nf-orbital collapse:** Goeppert Mayer in 1941 first pointed out DOC mechanism in his research paper titled rare earth and transuranic elements [64]. In heavy atoms with $f$ electrons the effective potential ($V_{\text{eff}}(r)$) is determined by the atomic central potential $V(r)$ and the centrifugal term $l(l + 1)/2r^2$ as $V_{\text{eff}}(r) = V(r) + l(l + 1)/2r^2$ where $l$ is orbital angular momentum [65]. The heavy atoms with $f$ electrons consist of two potential wells separated by a potential barrier. The outer potential well is dominated by the long-range coulomb potential and behaves asymptotically as $-1/r$ for neutral atoms. It is broad and shallow and can support an infinite Rydberg series of $nf$ bound states. The inner-well, on the other hand, is much narrower and deepens with increasing nuclear charge $Z$. For the lighter atoms, it is not deep enough to support any bound-state and all $nf$ wave functions reside in the outer-well. The first bound state of the inner-shell well appears near $Z = 58$, leading to sudden collapse of the $4f$ wave functions from the outer into the inner-shell forming an orbital which is spatially localized at nearly the same
radius as the orbital of the 4d state [46], and [65]. Similarly, since the attractive piece of the atomic potential is increased by the removal of outer-atomic electrons, many ionized system for which Z < 58 can also collapse 4f levels [46]. The mechanism is observed experimentally in the Xe, Ba, and La. But it fails in the Kr [46].

**Electron shake process**: The sudden change in atomic potential during the ionization of an inner-shell electron results in other atomic electrons to have a small probability to be excited from an outer-shell to an unoccupied bound-state (shake-up) or ejected into the continuum (shake-off) via a monopole transition. Nuclear decays such as α-particle emission, β decay, or electron capture also could induce the electron shake process. The shake-up states are located in the binding energies above the resonance states and the single-ionization threshold [66]. The monopole shake transition probabilities can be calculated with the sudden approximation. The calculated probabilities for Xe monopole excitations of 4d, 5s, 5p states after the creation of 1s and 2p inner-shells holes are presented in reference 67. The calculated nonzero electron shake-process probabilities confirm the electron shake process is one of the ionization mechanisms that take place during the interaction of pump-laser with Xe clusters.

The experimental evidences of the nf-orbital collapse and theoretical calculations of electrons shake process clearly shows dynamical orbital collapse mechanism is one of the possible mechanisms, which could take place during the laser-matter interaction process. As discussed earlier the excitations mechanism-taking place in XRIM is a general multiphoton multi-electron interaction, which involves the combined dynamics of MPI, orbital collapse, and correlated electron motion. Especially the dynamical orbital
collapse process enables the hollow-atoms having inner-shell vacancies to be produced under dynamical conditions in which the outer electrons of the systems are simultaneously and selectively arranged so that the over all electronic configurations formed exhibits the maximum radiative rate possible at the emitted wavelengths [46]. The ordered motions of electrons not only maximize the radiative emission but also enhance the coupling of pump-laser to the Xe clusters. An enhanced coupling involving an ordered motion of \(Z\) electrons would have three immediate consequences. They are (i) the effective coupling constant for the interaction becomes \(Z^2\alpha\), a value substantially increased from the customary magnitude \((\alpha \approx 1/137)\) (ii) a region of strong coupling can exist \((Z^2\alpha \geq 1)\) [68], and (iii) in analogy with ion-atom collisions, additional channels of ionization involving multiple electron ejection from inner-shells can become important [69]. The comparison of coupling constant for four fundamental interactions is presented below in the table 3.3.

<table>
<thead>
<tr>
<th>Interaction</th>
<th>Strength</th>
<th>Range</th>
<th>Systems</th>
</tr>
</thead>
<tbody>
<tr>
<td>Strong</td>
<td>(\sim 1)</td>
<td>(\sim 10^{-13}) cm</td>
<td>Nuclear matter</td>
</tr>
<tr>
<td>Electromagnetic in charge Particles</td>
<td>(\alpha \sim 1/137)</td>
<td>(\sim 1/r^2) (r \sim a_0)</td>
<td>Atoms, molecules, solids</td>
</tr>
<tr>
<td>Weak</td>
<td>(\sim 10^{-14})</td>
<td>(\sim 10^{-15}) cm (h/Mc)</td>
<td>Weak decay (\gamma)</td>
</tr>
<tr>
<td>Gravitational</td>
<td>(\sim 10^{-38})</td>
<td>(\sim 1/r^2)</td>
<td>Solar systems, galaxies</td>
</tr>
<tr>
<td>Channel radiative</td>
<td>(Z^2\alpha \sim 1 - 10)</td>
<td>(\lambda \gg a_0)</td>
<td>Ordered super-excited</td>
</tr>
</tbody>
</table>

Table 3.3: Comparison of basic forces.

A brief description of phenomena that involved with photons coupling with Xe atom is presented in reference 68, 70 and in chapter 4 along with the new experimental results produced in the D chamber with the help of calibrated higher resolutions diagnostics.
The fundamental purpose of this thesis is generation of ultrabright x-rays at laboratory-size and channeling the generated x-ray radiation in the forward-direction to produce directional x-ray source. The following sections discuss x-ray channeling mechanisms.

3.5 X-ray Channeling Mechanism

Series of two fundamental phenomena take place during the generation of ultrabright x-ray sources. They are (i) generation of ultrabright x-rays and (ii) non-linear propagation of electromagnetic x-ray radiation through the plasma that is generated. Part of x-ray source where x-ray radiations are channeled to propagate in the axial-forward direction is termed in this thesis as x-ray channel.

The x-ray channels could be defined as the stable compression and confinement of the spatial distribution of the x-ray pulses radiation into a narrow longitudinal plasma channel or dynamical waveguide, which is compensated for diffractive divergence over many Rayleigh lengths by virtue of self-induced regularization of the intrinsic refractive-index profile [71]. The experimental observations of XRIM provide convincingly enough evidences that the generated ultrabright x-ray radiations are channeled in the axial-direction of propagation of pump-laser beam. Further, previously developed models and theories of XRIM also predict that a self-guiding mechanism channels the generated ultrabright x-ray radiations in the axial-direction of propagation of the pump-laser beam. Previously proposed and developed theories and models of XRIM predicts that the laser-driven channels are hypothesized to support a degree of solitonic transport behavior, simultaneously stable in the space and time domains such a way the group velocity
dispersion balances self-phase modulations. The essence of the self-channeling mechanism is briefly discussed below in following paragraphs.

As mentioned earlier, there are already developed theories and models are available to explain the experimental observations of x-ray radiation propagation in plasma. Therefore, instead of developing new models and theories, previously developed theories and models are adopted to explain the experimental observation of x-ray radiation propagation in plasma.

At the end of the chapter 3, a simple theory of diffraction in finite-aperture is presented to estimate x-ray channel radius, which leads to estimate the intensity of directed ultrabright x-ray beam. The following section, x-ray self-channeling mechanism, briefly discusses the mechanism associated with the propagation of x-ray radiation in plasma.

3.5.1 Theoretical Aspect of X-ray Self-channeling Mechanism

The self-channeling mechanism begins with the creation of plasma column by ionization in the temporally early region of the pump-laser pulse and then high power pump-laser undergoes self-focusing.

The expression for the angular frequency ($\omega_L$) of the pump-laser and the angular-frequency of the plasma ($\omega_p$) are given below by equations 3.47, and 3.48 respectively.

$$\omega_L = \frac{2\pi c}{\lambda_L} \quad (3.47)$$

$$\omega_p = (4\pi^2 N_e / m_e)^{1/2} \quad (3.48)$$

Where $N_e$ is initial unperturbed electron density, and all the notations have their previously described meanings.
During the propagation of the pump-laser through the fully ionized plasma ponderomotive force \( F_{pon} = -e^2 \nabla^2 E / 4m_0 \omega_L^2 \) will displace electrons from the high-intensity-zone. The ions, which are relatively heavier than electron in the plasma, are regarded as motionless for a short time due to their substantially greater inertia during the propagation of the pump-laser pulses through the underdense-plasma. The required time, termed as characteristic-time \( (\Delta t_i) \), for ions to move on a fraction of the focal-spot radius \( (r_{focus}) \) is estimated using equation 3.49 presented below [72].

\[
\Delta t_i = \frac{2\pi r_{focus} c}{e} \sqrt{\frac{m_e m_p \epsilon_0 c^3}{2I_L \lambda_L^2}}
\]

Where \( m_p \) is the proton mass and all other notations have their usual meaning.

The estimated characteristic time \( (\Delta t_i) \) for the experimental conditions for generation of ultrabright x-ray source is approximately 600 fs. It is clear from the estimation that the ions are motionless for such shorter time and the electrons and ions are segregated within the characteristic time. In this thesis, the electrons and ions segregation mechanism due to the ponderomotive force is termed as charge-separation or cavitation mechanism.

The charge-separation mechanisms segregate electrons and ions and form a dielectric waveguide similar to a capillary tube. Since the wall of the dielectric waveguide is made out of electrons, the skin-depth \( (\delta) \) (penetration of electromagnetic wave to a distance before attenuating to zero) for the x-ray electromagnetic radiation is higher at the wall of the dielectric waveguide. Therefore, the generated x-ray radiation will be self-trapped inside the dielectric waveguide and will be channeled to propagate in the axial-direction of pump-laser beam. The Thomson scattering (scattering of pump-
laser photons from free electrons) experimental data, presented in the following chapter 4, clearly shows evacuation of electrons at the center of pump-laser beam and formation of x-ray self-channel.

Refractive-index spatial fluctuation within the plasma medium, through which the x-ray radiation propagates, determines x-ray channeling mechanism and the morphology of the propagating x-ray radiations. The influence of the spatial variation of refractive index, arising from the nonlinear response of the dielectric properties of the propagating plasma medium, depend on three parameters (i) electron density \( N_e \), (ii) electron mass \( m_e \) (iii) induced electron-ion dipole. Sections presented below briefly discuss, one by one, the influence of these three plasma parameters on the x-ray channeling mechanisms.

The following section briefly presenting how the cavitation (or charge-separation) mechanism alters electrons density \( N_e \) and hence modify index of refraction of the plasma medium \( n_p \) through which ultrabright x-ray radiation propagates.

### 3.5.1.1 Influence of Electron Density Variation on X-ray Channeling Mechanism

As presented below in equation 3.50 the refractive-index of plasma medium \( n_p \) could be express in terms of plasma frequency \( \omega_p \) and laser frequency \( \omega_L \).

\[
n_p = \left( 1 - \frac{\omega_p^2}{\omega_L^2} \right)^{1/2}
\]  

(3.50)

As discussed earlier, during the cavitation mechanism the ponderomotive-force expels electrons from the center of the pump-laser to annular region while the ions remain stationary for short time at the center of the pump-laser. The segregation of
electrons and ions lead to a charge imbalance creating an electrostatic-field attracting electrons back to the center of the pump-laser beam. The balance between those two forces, radial electrostatic-force and ponderomotive-force creates electrons density gradient along the radial-direction.

According to the expression for the plasma frequency $\omega_p = (4\pi e^2 N_e / m_e)^{1/2}$ and the equation 3.50, creation of the radial electron density ($N_e$) gradient in the radial-direction alters the index of refraction of the plasma-medium in such a way that the refractive-index of the plasma decreases along the radial-direction. The radially inverted refractive-index of the plasma behaves like gradient index-optics, such as positive-lens, to the propagating ultrabright x-rays and self-focuses x-ray radiations.

As discussed above the **cavitation** mechanism creates on-axis electron density ($N_e$) minima and produces radially dependent focusing index-guide. However, in the other hand the tunneling and multi-photon ionization in the focal-region has the opposite defocusing effect by producing higher electron densities. The balance between the defocusing and self-focusing effects produces multi-foci points within the x-ray channel during the propagation of ultrabright x-ray radiation through plasma medium. The multi-foci and de-foci spatial locations within the x-ray channels are clearly identified experimentally with the helps of high-resolution aperture x-ray camera and Thomson free electrons scattering diagnostics. The detailed data are presented in the following chapter 4.

The influence of the second plasma parameter, electron mass ($m_e$), on the x-ray channeling mechanism is discussed in the following section.
3.5.1.2 Influence of Relativistic Electron Mass Variation on X-ray Channeling

Mechanism

As discussed earlier, the ponderomotive force drives plasma electrons and accelerates them. The expression for the quiver velocity \( V_q \) of electrons under the influence of linearly-polarize laser-light-field due to the ponderomotive force is presented below in the equation 3.51.

\[
V_q = \frac{eE_L}{m_e \omega_L}
\]  

(3.51)

Where all the notations have their previously defined meanings.

The electron’s quiver velocity \( V_q \) is estimated by using above equation 3.51 for the pump-laser parameters (presented in the table 3.1) at the focal-plane. The estimated electron’s quiver velocity is \( 17 \times 10^6 \) m/s. Since the quiver velocity is in the order of velocity of light, it is in the relativistic regime and mass of the electrons depend on its velocity.

In the theory of relativity, the relativistic electron mass \( m_{re} \) is expressed by equation 3.52 presented below.

\[
m_{re} = \frac{m_e}{\sqrt{1 - \frac{V_q^2}{c^2}}}
\]  

(3.52)

Where \( m_e \) is the electron’s rest-mass, \( V_q \) is the quiver velocity of electrons, and \( c \) is the velocity of light.

According to the equation 3.52, the electron’s quiver motion, under the influence of the pump-laser field, increases its relativistic electron-mass \( m_{re} \). Increased relativistic-
mass \( m_{re} \) decreases plasma angular-frequency as the plasma angular-frequency \( \omega_p \) 
\( (= (4\pi e^2 N_e / m_e)^{1/2}) \) is inversely proportional to the relativistic electron-mass \( m_{re} \).

According to the expression for the refractive index of plasma \( n_p \), presented in the equation \( 3.50 \), the decreased angular-frequency \( \omega_p \) of the plasma-medium will increase the refractive-index of the plasma through which the ultrabright x-ray radiation propagates. The elevated refractive-index of plasma due to the electron’s quiver motion will act again as gradient-index-optics and will self-focus the propagating ultrabright x-ray radiations. In this thesis, the self-focusing phenomenon due to the relativistic electron’s quiver motion is termed as \textit{relativistic self-focusing}.

The third mechanism, which influences the x-ray channel formation and propagation is the nonlinear response arising from the induced dipoles of the ions. However, this mechanism is generally small and negligible compare to first two mechanisms cavitation and relativistic self-focusing.

In reality, the x-ray channel formation and propagation is a very complicated mechanism. The two mechanisms \textit{cavitation} and \textit{relativistic self-focusing} are not enough to explain experimental observations completely. The references 73, 74, and 75 briefly discuss the available models and self-channeling theories.

**3.5.3 Estimating X-ray Self-channel Radius**

The \textit{cavitation} mechanism creates dielectric waveguide and channeling x-ray radiation by trapping it and self-focusing it. However, in the other hand the x-ray channels are defocused at the end of the waveguide by various mechanisms such as diffraction from the finite-aperture and refraction by the transverse in-homogeneities in
the electron density due to ionization mechanisms and the in-homogeneities of the pump-laser beam profile. As discussed earlier, the scenario of channeling and defocusing is similar to propagation of radiations through a dielectric waveguide. The theory of propagation of radiation through dielectric waveguide is very complicated. In this thesis, the complicated theories of waveguide are not presented. However, it is assumed that the defocusing at the end of the dielectric waveguide or x-ray self-channel is caused by the diffraction of x-ray radiations when it passes through finite-aperture as shown below in diagram 3.3.

![Diagram 3.3: Diagrams illustrates general x-ray source morphological image captured by aperture x-ray camera. In general x-ray source morphological images has converging cone shape entrance-zone and diverging cone shape exit-zone and in between them a dark-zone termed in this thesis x-ray self-channel. Diffraction at the end of dielectric waveguide is also shown in the diagram.](image)

The simple diffraction equation, which relates the size of the aperture ($2d_{cha}$) and the diffraction angle ($\theta$) to the x-ray radiation wavelength ($\lambda_x$) is presented below by equation 3.54.

$$2d_{cha} \sin \theta = \lambda_x \quad (3.54)$$

The diffraction-angle ($\theta$) will be estimated, in the following chapter 4, from aperture x-ray camera and Thomson scattering channel images for the particular x-ray
radiation wavelength ($\lambda_x$). The x-ray channel-radius ($r_{ch}$) will be estimated using the equation 3.54 for the experimentally estimated parameters ($\theta$, and $\lambda_x$) values.

In reality, all the pump-laser energy falls into Xe/ Kr clusters is not converted into x-ray radiation. In a typical experiment, the pump-laser energy is dissipated into (i) motion of the electrons, (ii) ionization of the gas atoms, (iii) generation of harmonic radiation, (iv) production of inverse bremsstrahlung, (v) Compton scattering, and (vi) other amplitudes of nonlinear scattering. However, in this thesis, the generated x-ray energy ($E_x$) is measured in three different ways. The details of measurements are presented in the following chapter 4. The pulse-width ($\tau_x$) of the x-ray pulses are estimated from spectral feature collected from axial-von Hámos single-crystal spectrometer and presented in the following chapter 4. The measured x-ray energy ($E_x$) and the estimated pulse-width ($\tau_x$) leads to estimate the x-ray radiation power ($P_{ch}$) trapped within the x-ray channel and intensity of channel. The ratio of pump-laser power to the x-ray power trapped inside the x-ray channel will be estimated in the chapter 4 in order to determine the power compression during the generation of laboratory-scale ultrabright x-ray sources.

The chapter 3, theoretical background and estimations, is the tool to understand the single-shot data recorded in the generation of ultrabright x-ray source experiments. The following chapter 4, results and discussion, presents experimental results along with discussions and leads to conclusions in final chapter 5.
4. RESULTS AND DISCUSSIONS

4.1 Introduction

Fundamental goals of research works presented in this thesis are generation of ultrabright laboratory-scale x-ray sources in the kiloelectron Volt (keV) wavelength regime and optimizing the realized x-ray sources. The section 2.3 of chapter 2 presented the experimental methods and apparatus for generation of laboratory scale ultrabright x-ray sources. The chapter 3 is presenting a tool, theoretical framework, to understand the physics behind the x-ray source and experimental observations presented in chapter 4.

Chapter 4, results and discussions, is presenting experimental results along with discussions. In the chapter 4, recorded single-shot experimental results for Xe and Kr x-ray sources are presented separately in two primary sections 4.2 and 4.3. The sections 4.2 and 4.3 are further divided into three categories (i) triplet-aperture x-ray camera, (ii) Thomson scattering, (iii) axial and transverse-spectrometers.

The subsection of triplet-aperture x-ray camera is presenting wavelength range of the x-ray source, various morphological patterns of the x-ray source. At the end of this subsection, energy of the realized x-ray sources is estimated from the single-shot x-ray image capture by the triplet-aperture x-ray camera.

The subsection of Thomson scattering (TS) diagnostics is discussing propagation dynamics of generated x-ray radiations. Information extracted from x-ray source images captured by the TS are leading to estimate length and diameter of x-ray self-channels.

The subsection of axial and transverse-von Hármos single-crystal spectrometers is presenting recorded spectroscopic information during the interaction of pump-laser with
target-clusters. At the end of this subsection, x-ray energy flow in the axial-direction is estimated from the data recorded by the single-shot axial-spectrometers. The axial-energy estimation is leading to estimate brightness of realized x-ray sources.

As presented in the chapter 2 a hybrid Double Gas Sonic Nozzle (DGSN) is custom designed to spray primary single target-gas at center and low-Z secondary gas in the primary target-gas annular area to optimize target-cluster density while reducing x-ray attenuation during its propagation through unionized target-gas column. However, in all experiments, except experiments presented in chapter 4.4, the DGSN is used as a single-gas sonic-nozzle to spray only primary target-gas to produce target-clusters. Chapter 4.4 verifies the operation of DGSN and its design purpose of optimizing x-ray source. Single-shot axial-Xe $M$-shell x-ray energies are estimated with (DGSN sprays only primary gas) and without (DGSN sprays both primary and secondary gas) operation of DGSN.

4.2 X-ray Source Generation from Xe Hollow-atom States

In this section 4.2, Xe x-ray source generated from Xe hollow atomic states are discussed. Figure 4.1 presents below a sample single-shot image of realized x-ray source recorded by the calibrated triplet-aperture x-ray camera.
Figure 4.1: Single-shot Xe x-ray source image captured by calibrated triplet-aperture x-ray camera.
(a) Xe $M/L$-shell x-ray source image captured by 50 µm aperture along with shaded surface image.
(b) Xe $L$-shell x-ray source image captured by 100 µm aperture along with shaded surface image. Magnification factor of the aperture camera is ignored in the picture.
Red ellipses in the figure 4.1 (a) is termed as entrance and exit-zones.
As shown in the above figure 4.1 (a) and diagram (3.3), right-half cone-shaped portion of the x-ray source will be referred, in this thesis, as exit-zone and left-half cone-shaped portion of the x-ray source will be referred as exit-zone during the discussions.

As presented in the chapter 2, the aperture x-ray camera has triplet-apertures with 100 µm, 50 µm, and 25 µm aperture sizes. The 25 µm and 50 µm apertures are covered with a 10 µm thickness Be foil for suppressing all radiations longer than the interested Xe M/L-shell x-ray radiations from x-ray source. The 100 µm aperture is covered with 7.5 µm thickness Ti foil in addition to the 10 µm thickness Be foil for suppressing all the radiation longer than Xe L-shell x-ray radiations from the same x-ray source. Figure 4.2 below presents transmission graphs of the filters. The triplet-aperture x-ray camera is aligned, as shown in the figures 2.19 and 4.6, in the transverse-direction to the propagating pump-laser beam.

The filter transmission graph presented below in figure 4.2 and the image of x-ray source presented in the figure 4.1 confirms that the interaction of intense 248.6 nm pump-laser with Xe cluster molecules produce x-ray radiation in the x-ray wavelengths regime.
At this point, this thesis, make it clear that Xe _M-shell_ data recorded by the aperture x-ray camera and von Hámos single-crystal spectrometers contain Xe _M-shell_ and Xe _L-shell_ x-ray radiations. In the other hand, Xe _L-shell_ data recorded by the aperture x-ray camera and von Hámos single-crystal spectrometers contain dominated Xe _L-shell_ x-ray radiations.

Brief analysis of x-ray source images captured by the triplet-aperture x-ray camera is presented in following section 4.2.1. In additions to the triplet-aperture x-ray camera diagnostic, three other diagnostics (TS, axial and transverse-von Hámos single-crystal spectrometers) are visualizing the x-ray source. Recorded raw-data by these three diagnostics are presented below in figure 4.3.
Experimental raw-data images at a glance

Following sections are analyzing experimental single-shot data recorded by all four diagnostics.

| UV energy | 317 mJ |
| Xe temperature | 292K |
| Xe pressure | 164 psig |

Shot No. : 375  
Exp. date: 08/17/2012

Figure 4.3: Experimental raw data at a glance. Picture presented in the left bottom corner is presenting x-ray source image captured by the aperture x-ray camera, which is overlaid with DGSN. The red scale shows the dimension of the DGSN. Concentric circles show perimeters of DGSN orifices. The right top picture is presenting line focus image captured by transverse-von Hámos single crystal spectrometer. The right center picture is presenting line focus image captured by axial- von Hámos single crystal spectrometer. The right bottom picture is presenting x-ray source image captured by three times magnified Thomson scattering diagnostics optical setup.
4.2.1 Analyzing Single-shot Data Recorded By Triplet-aperture X-ray Camera

This section is briefly analyzing the single-shot data recorded by the triplet-aperture camera. All the images captured by the triplet-aperture x-ray camera during the experiment are carefully analyzed and results are presented in following sections. The following section, 4.2.1.1, is discussing experimentally observed different-types of morphological structures of the Xe x-ray source.

4.2.1.1 Different Types of Xe X-ray Source Morphological Structures

Triplet-aperture x-ray camera single-shot data shows that x-ray source morphological structure types vary shot by shot. Figure 4.4 below presents six commonly observed morphological patterns of the realized Xe x-ray sources by 50 µm aperture camera.
Figure 4.4 (a): Experimentally observed first type x-ray source morphological structure. The figure shows an entrance-zone and exit-zone. Between entrance and exit-zones the triplet-aperture x-ray camera, at its best resolution, does not observe x-ray radiations.

Figure 4.4 (b): Experimentally observed second type x-ray source morphological structure. The figure shows an entrance-zone and exit-zone and a radiation between entrance and exit-zones.
Figure 4.4 (c): Experimentally observed third type x-ray source morphological structure. The figure shows an x-ray self-channel formation instead of entrance-zone. The exit-zone has wider radiation volume and it is bifurcated into two.

uv energy: 319 mJ
Xe temperature: 292 K
Xe pressure: 148 psig

Shot No.: 531
Exp. date: 08/17/2012

Figure 4.4 (d): Experimentally observed fourth type x-ray source morphological structure. The figure shows weak entrance-zone. The exit-zone is bifurcated into two. A weak x-ray self-channel formation is observed between entrance and exit-zones.

uv energy: 293 mJ
Xe temperature: 292 K
Xe pressure: 147 psig

Shot No.: 543
Exp. date: 08/17/2012
There are various experimental parameters deciding the x-ray sources morphological structure type. Temporal and special pump-laser beam profiles and DGSN
poppet actuate timing are two dominating parameters, which decide x-ray source morphological structure types. It was experimentally observed that changing round-trip timing of regenerative-amplifier of Hurricane seeding-laser and changing the DGSN poppet actuating-timing (effective timing range is 675 ± 25 µs) switching the x-ray source morphological structure type from one type to another type. The performed experiments for this thesis had complete control of selecting desired x-ray source morphological type during the experiment. Complete understanding reasons for various x-ray source morphologies demand recording single-shot data of temporal and spatial profile of pump-laser. Such recorded single-shot data set could perceive hidden information behind the various x-ray source morphological types. This thesis is not discussing detail of such study but it is the first future objective of this thesis.

4.2.1.2 Coupling of Xe M-shell radiation to Xe L-shell radiation

Figure 4.5, presented below, shows simultaneously recorded four single-shot triplet-aperture x-ray camera Xe M-shell and Xe L-shell emission images.
uv energy: 290 mJ
Xe temperature: 292 K
Xe pressure: 162 psig

Shot No.: 397
Exp. Date: 08/17/2012

uv pulse-width: ~ 400 fs

$D_{\text{source}} \sim 400 \text{ µm}$
$I_{uv} = 14 \times 10^{16} \text{ W/cm}^2$

Figure 4.5 (a)

uv energy: 349 mJ
Xe temperature: 292 K
Xe pressure: 155 psig

Shot No.: 458
Exp. Date: 08/17/2012

Figure 4.5 (b)
Figure 4.5: Xe M-shell emissions volumes are matching with Xe L-shell emission volume for arbitrarily selected four different experimental shots confirm that the Xe M-shell radiation is coupled to the Xe L-shell radiation.
The figure 4.5 clearly shows that the Xe \textit{M-shell} emission is spatially overlapping with Xe \textit{L-shell} emission. As shown in the figure 4.5 (a) the breadth of the x-ray source at the entrance-zone is \( \sim 400 \) \( \mu \)m. The intensity of the pump-laser (\( I_{\text{p}} \)) at this particular point is \( \sim 1.4 \times 10^{17} \) W/cm\(^2\) (shown in the figure 4.5 (a)). The intra-cluster-model, presented in the section 3.4.2.5 of chapter 3, predicts that the pump-laser intensity \( 1.4 \times 10^{17} \) W/cm\(^2\) is enough for the direct production of Xe \textit{M-shell} emission. However, at this intensity level, the UV pump-laser is not capable of producing direct Xe \textit{L-shell} emission. Therefore, the experimentally observed broad zone of Xe \textit{L-shell} emission predicts that the energetic Xe \textit{L-shell} emission is excited directly by the strong Xe \textit{M-shell} signal. The observed overall nonlinear absorptive process could be express by the formula presented below in the equation 4.1.

\[
n \gamma(M) \rightarrow Xe^*(L) \rightarrow \gamma(L)
\] (4.1)

In the above equation 4.1, for direct production of Xe \textit{L-shell} emission from the unexcited Xe cluster molecules the minimum value of \( n \) should be 5 based on the energies of the states involved [70].

The coupling of Xe \textit{M-shell} emission to the Xe \textit{L-shell} emission is a nonlinear coupling and the two observed nonlinear couplings (dispersive and absorptive), as illustrated above in figures 4.5, can be explained by the nonlinear Kramers-Kröning relation [70]. The experimental observation of the nonlinear coupling of Xe \textit{M-shell} emission to the Xe \textit{L-shell} emission led to a research paper presented in reference 70 and brief discussion of the nonlinear coupling of Xe \textit{M-shell} emission to the Xe \textit{L-shell} emission is presented in reference 70.
Complete theoretical and experimental studies are necessary to understand how the Xe $M$-shell x-ray radiations excite the Xe L-orbits. One of the proposed ionization mechanisms is MPI mechanism. As discussed earlier, $5(=n)$ Xe $M$-shell photons could excite the Xe $L$-shell in the MPI fashion. A necessary condition for the MPI mechanism to take place is that the Xe $M$-shell x-rays should have to have coherent nature. The other proposed ionization mechanism is that the Xe $M$-shell x-rays, as presented in the tunneling ionization mechanism, modify the Xe potential well and cause Xe L-orbit to ionize. The second case also demands the coherent nature of Xe $M$-shell x-rays. Therefore, regardless how the Xe $M$-shell x-rays ionize the Xe L-orbit, Xe $M$-shell x-rays should have to have some degrees of coherent nature, which is one of the laser-like properties. Further, the single-shot triplet-aperture x-ray camera images presented in figures 4.4 and 4.5 clearly show that the Xe $M$-shell x-ray radiations are under-going self-focusing during its propagation. The propagating Xe $M$-shell x-ray radiations should satisfy two vital conditions for the self-focusing mechanisms taking place during the propagation of Xe $M$-shell x-rays. The conditions are (i) power of propagating Xe $M$-shell radiation should exceed the critical-power and (ii) the propagating Xe $M$-shell radiation should have to have coherent nature. Therefore, the observed self-focused Xe $M$-shell radiation in the single-shot triplet-aperture x-ray camera images presented in the figures 4.4 and 4.5 reiterate that realized Xe $M$-shell is having some degrees of coherent nature. As discussed in the section 3.4.2.6 of chapter 3, one of the possible ionization mechanisms, which produces Xe $M$-shell x-ray radiations is ordered driven electron motions in the attosecond regime (less than electrons de-phasing time). This type of mechanism has high chances of producing coherent x-rays.
Furthermore, the theoretical works presented in the section 3.4.2.5 of chapter 3 predicted that the minimum intensity for the production of Xe \(L\)-shell emission with 248.6 nm radiation is in the order of \(10^{18}\) W/cm\(^2\) (figure 3.9). The new experimentally observed nonlinear coupling of Xe \(M\)-shell to the Xe \(L\)-shell emission mechanism reduces this minimum intensity for the production of Xe \(L\)-shell emission by a factor of \(~10\) and favors the production of deep inner-shell vacancies, which could lead to realize shortest \textit{coherent} intense radiations.

### 4.2.1.3 Estimating Xe X-ray Source Energy via Single-shot Image Captured by Calibrated Aperture X-ray Camera

In this section, estimation of single-shot Xe x-ray source energy from the x-ray source image captured by the single-shot triplet-aperture x-ray camera is presented. Principles of aperture x-ray camera and the alignments details are present in the section 2.3.3.1 of chapter 2.

In a typical triplet-aperture x-ray camera diagnostics setup, x-ray radiation from the x-ray source passes through single-point aperture which is covered with appropriate filter to attenuate all the radiation longer than desired radiation and projects an inverted image on CCD chip of the PI camera located a certain distance from the aperture. Figure 4.6 illustrates the optical-ray diagram of the triplet-aperture x-ray camera.

The PI CCD chip has an epitaxial layer of silicon, which eject photoelectrons for the incident of x-ray radiations. An electrical capacitor is built-up with each pixel to accumulate photoelectric charge proportional to the radiation intensity at that location. In the triplet-aperture x-ray camera diagnostics, x-ray source image projected through an
aperture onto the photoactive region, causing each capacitor to accumulate an electric charge proportional to the light intensity at that location. A control-circuit causes each capacitor to transfer its contents to its neighbor (operating as a shift register). The last capacitor in the array dumps its charge into a charge amplifier, which converts the charge into a voltage. By repeating this process, the controlling circuit converts the entire contents of the array in the semiconductor to a sequence of voltages and image captured by the aperture camera will be retrieved.

According to the manufacture specification, the PI CCD chip requires 3.65 eV average incident photon-energy to release a photoelectron from CCD chip Si material and 25 of such photoelectrons will be converted to 10 gray-level counts (or pixel voltage) \([41]\). Therefore, if one knows total gray-level counts in an x-ray source image captured by the aperture diagnostics, it is possible to estimate total number of electrons ejected from Si and hence the total photons energy incidents on the CCD chip. The total x-ray photons energy propagating through aperture and incident on the CCD chip is equal to total gray-level counts of the x-ray image times 2.5 times 3.65 eV. The equation 4.2, presented below, is presenting expression for total incident x-ray energy on the PI CCD chip \((E_{ccd})\).

\[
E_{ccd} = \text{Total gray-level of x-ray image} \times 2.5 \times 3.6 \text{ eV} \quad (4.2)
\]

The realized x-ray source is emitting x-ray photons into \(4\pi\) solid angle as shown below in the figure 4.6. However, the triplet-aperture x-ray camera is collecting x-ray photons only within the solid angle shown in the figure 4.6 by the dotted lines. Therefore, the total number of photons emitted by the x-ray source is equal to number of photons incident on the PI CCD chip times \(4\pi\) divided by aperture solid angle.
If the total number of photons emitted by the x-ray source is \( N_{Tot} \), the total number of photons leaving sphere defined in the figure 4.6 is also equal to \( N_{Tot} \). Therefore, the number of photons emitted by the x-ray source (\( N_{Tot} \)) could be express in terms of number of photons incident on the PI CCD chip (\( n_{ccd} \)), aperture radius (\( r_{Ape} \)), and the sphere radius (\( R_{Spe} \)). The expression is presented below by equation 4.3.

\[
N_{Tot} = n_{ccd} \times \frac{4R^2}{r_{Ape}^2} \quad (4.3)
\]

Figure 4.6: Triplet-aperture x-ray camera experimental setup. The figure shows the x-ray source emits radiation in all \( 4\pi \) direction. The PI image-plane is rotated 60 degrees from the XZ plane and aperture is viewing the x-ray source in the transverse direction to the x-axis.

The x-ray source is producing different number of photons at distinct energies (or distinct x-ray wavelengths). An expression for total energy of photons (\( E_{ccd} \)) incident on PI CCD chip is presented below in the equation 4.4.
\[ E_{ccd} = \sum_{i=1}^{N_{Pho}} n_i E_i \]  

(4.4)

Where \( n_i \) is total number of photons with particular energy \( E_i \), \( N_{Pho} \) is total number of different energetic photons, and \( \sum_{i=1}^{N_{Pho}} n_i = n_{ccd} \).

A mathematical expression, which is similar to the equation 4.4 could be derived for expressing total number of x-ray photons emitted by the Xe x-ray source. In such expression, \( E_i \) remains same while number of photons in a particular energy \( E_i \) will be increased by the factor of \( 4R^2/r_{Ape}^2 \). Therefore, the total the x-ray source energy (\( E_{Tot} \)) could be expressed by an equation 4.5 presented below.

\[ E_{Tot} = \sum_{i=1}^{N_{Pho}} n_i \times \frac{4R^2}{r_{Ape}^2} \times E_i \]  

(4.5)

\[ \Rightarrow E_{Tot} = \frac{4R^2}{r_{Ape}^2} \times \sum_{i=1}^{N_{Pho}} n_i E_i = \frac{4R^2}{r_{Ape}^2} \times E_{ccd} \]  

(4.6)

In the equation 4.5, \( R \) is equal to 8.5 cm, \( r_{Ape} \) is equal to 50 µm, and the \( E_{ccd} \) could be estimated from the equation 4.2.

The total x-ray source energy is estimated using the equation 4.6 for the experiment performed on August 17, 2012 and presented below in figure 4.7.
Figure 4.7: Graph of Xe M-shell and Xe L-shell x-ray energy versus shot number. The average Xe M-shell x-ray energy is 41 µJ and Xe L-shell x-ray energy is 3 µJ. In the energy estimation all the factors, which attenuate x-rays during its propagation to PI CCD chip are ignored.

The Xe M-shell data-set and Xe L-shell data-set presented in the above figure 4.7 clearly shows strong correlations between Xe M-shell emission and Xe L-shell emissions. Xe M-shell data point distributions and Xe L-shell data point distributions in the figure 4.7 are following similar pattern, which reconfirms that the Xe M-shell and Xe L-shell are coupled together.
Single-shot $L$-shell x-ray energy versus Xe $M$-shell x-ray energy graph is plotted and presented conforms that the Xe $L$-shell is depend on Xe $M$-shell x-ray energy and supports coupling of Xe $M$-shell emission to Xe $L$-shell emission. The ratio of Xe $M$-shell x-ray energy to Xe $L$-shell x-ray energy is about 16.

![Graph](https://example.com/graph.png)

**Xe L-shell versus Xe M-shell energy**

- UV average energy: 252 mJ
- Xe average pressure: 160 psig
- Xe temperature: 293 K
- Exp. date: 08/17/2012

Figure 4.8: Graph of Xe $L$-shell x-ray energy versus Xe $M$-shell x-ray energy. The ratio of Xe $M$-shell x-ray energy to Xe $L$-shell x-ray energy.

In the Xe x-ray energy estimations, it is assumed that x-rays are not attenuated during the propagation of x-rays photons from x-ray source to the PI camera CCD chip. However, as presented in the section 2.3 of chapter 2 and in the end of chapter 4,
unionized target-gas molecules stays in the vicinity of target area after generation of x-rays and attenuate propagating x-rays. Transmission graph for 2.65 mm Xe gas column is presented in figure 4.51 at the end of chapter 4 shows that Xe M-shell x-ray are attenuating more than 80% during the propagation of Xe X-rays through 2.65 mm Xe gas column.

In addition to the unionized gas column the filter through which the x-ray radiations passes through is also attenuating reasonable amount of x-ray radiation during their propagation. The transmission graphs for M-shell and L-shell, presented in the figure 4.2, clearly shows that the M-shell x-ray radiations are attenuated at least factor of 30 during the propagation.

Further, it is assumed, in the estimation, that the x-ray source is emitting x-ray photons uniformly in all $4\pi$ directions. If the x-ray self-channeling formation takes place during the propagation of x-ray radiation, the total number of x-ray photons in the transverse-direction, obviously, will be smaller than the total number of x-rays photons in the forward-direction and hence the x-ray energy estimations give lower value for the total Xe x-ray source energy estimations. Energy flow in the forward axial-direction is estimated from axial-spectrometer and presented in section 4.2.3. Estimated x-ray energy flow in the axial-direction is higher than estimated x-ray energy flow in the transverse-directions confirming existence of x-ray self-channel formation during the generation of Xe x-ray sources.

In the Xe x-ray source, energy estimations above discussed x-ray attenuation factors are ignored. Therefore, x-ray source energy estimated from the Xe x-ray source image captured by the triplet-aperture x-ray camera gives lower-bound value and the
actual x-ray source energy should be considerably higher than the estimated value. Upper-bound value for Xe x-ray is predicted hypothetically as follows. The Xe \textit{M-shell} wavelength range is 0.95-1.6 nm (0.8- 1.2 keV). The energy of a single pump-laser photon is \( \sim 8 \times 10^{-19} \) J (wavelength 5 eV). If the average energy of the pump-laser inside the D chamber is 300 mJ, the total number of pump-laser photons available for the excitation is in the order of \( 10^{18} \). The average energy to ionize Xe 3d orbit to produce \textit{M-shell} is in the order of 1500 eV (2.4 \( \times 10^{-16} \) J). Therefore, a simple estimations, without considering complicated ionization mechanisms, show that 300 pumping photons has to interact with a single Xe \textit{M-shell} 3d orbit to generate a single \textit{M-shell} x-ray photon. If one assumes that the all the available pumping photons are only producing Xe \textit{M-shell} x-rays from 3d orbit, the total number of Xe molecules with ionized 3d orbit is \( \sim 10^{15} (=10^{18}/300) \). If the 3d ionized Xe ions produce Xe \textit{M-shell} x-ray with average energy 1000 eV (1.6 \( \times 10^{-16} \) J), the total energy of the Xe \textit{M-shell} x-ray produced by the interaction is in the order of 160 mJ (\( 10^{15} \times 1.6 \times 10^{-16} \) J). However, in reality the pump-laser photons are not only interacting the Xe \textit{M-shell} orbits to produce hollow atom states but also dissipated into motion of the electrons, generation of harmonic radiation, production of inverse bremsstrahlung, Compton scattering, and other amplitudes of nonlinear scattering. Therefore, above estimated value, 160 mJ, is an upper-bound ideal value for the realized Xe \textit{M-shell} x-ray source. The triplet-aperture x-ray camera estimation gives lower-bound average value of 41\( \mu \)J for the realized Xe x-ray source energy, which is \( \sim 4 \times 10^{3} \) times smaller than the ideal value for Xe \textit{M-shell} x-ray source energy.
Thomson Scattering (TS) is another diagnostics capture the image of the realized x-ray source. Figure 4.9, presented below, compares four different x-ray source morphological type images captured by the triplet-aperture x-ray camera with images captured by the TS optical setup. The principles and details of the TS alignment along with alignment procedures are presented in the section 2.3.3.3.1 of chapter 2.
Figure 4.9 (a): Comparison of aperture x-ray camera image with 3 times magnified TS image.
The TS image shows the channel formation however, the resolution of aperture x-ray camera is not enough to show channel formation.

- **50 µm aperture x-ray camera image**
- **Three times magnified Thomson scattering image**

*uv laser direction*  
Xe M/L-shell radiation

- uv energy: 356 mJ  
- Xe temperature: 292 K  
- Xe pressure: 154 psig

Shot No: 470  
Exp. date: 08/17/2012
Figure 4.9 (b): Comparison of aperture x-ray camera image with 3 times magnified TS image. The TS image shows the self-channel formation clearly. However, the resolution of aperture x-ray camera is not enough to show channel formation.
Figure 4.9 (c): Comparison of aperture x-ray camera image with 3 times magnified Thomson scattering image. The TS image shows clearly the self-channel formation however, the aperture x-ray camera barely shows channel formation.
50 µm aperture x-ray camera image

Three times magnified Thomson scattering image

uv energy: 340 mJ  
Xe temperature: 292 K  
Xe pressure: 152 psig  
Shot No: 485  
Exp. date: 08/17/2012

Figure 4.9 (d): Comparison of aperture x-ray camera image with 3 times magnified TS image. The TS image shows the self-channel formation. However, the resolution of aperture x-ray camera is not enough to shows channel formation.
The above figure 4.9 clearly shows the x-ray source images captured by the TS are having similar features compare to the image captured by the triplet-aperture x-ray camera. Further, the images clearly show that the TS are having higher resolution compare to the triplet-aperture x-ray camera. In all four pictures of the figure 4.9, TS images show the x-ray self-channel formation. Even thou the triplet-aperture x-ray camera images are magnified 4 times by IDL software to perceive x-ray channel formation, the images captured by the triplet-aperture x-ray camera are barely showing the x-ray self-channel formation in the all four images of the figure 4.9. Since the TS optical setup provides higher resolution compare to the triplet-aperture x-ray camera, in this thesis TS images will be used, instead of x-ray aperture image, to study the x-ray source morphology. The following section 4.2.2 is analyzing the single-shot data recorded by the Thomson scattering diagnostics.

4.2.2 Analyzing Single-shot Data Recorded by Thomson Scattering Diagnostics

The Thomson scattering is scattering of pump-laser from free-electrons. Experimental aspect of the TS is presented in the section 2.3.3.3.1 of chapter 2. Since the TS provides higher resolution compare to all other diagnostics, it is used as a tool to understand propagation dynamics and morphological structure types of the x-ray radiations. The following section is characterizing x-ray self-channel formation, which channels generated x-ray radiations into a dielectric waveguide.
4.2.2.1 Characterizing X-ray Self-channel Formation by Thomson Scattering

As discussed previously the TS setup, compare to the triplet-aperture x-ray camera, perceive hidden information such as x-ray self-channel formation during the generation of ultrabright laboratory-scale x-ray source. In this section the TS is used as a tool to characterize the x-ray self-channel formations.

The theory of self-channeling, presented in the section 3.5 of chapter 3, predicts that the generated x-ray radiations from interaction of pump-laser with Xe cluster molecules will be trapped inside dielectric-waveguide and channels x-ray radiations in the axial-direction to the direction of propagation of the pump-laser. All four TS images presented in the figure 4.9 give clear evidences for the x-ray self-channel formation in the experiments for generation of laboratory scale ultrabright kilovolt Xe x-ray source.

For the purpose of estimating self-guided channel length, the on-axial intensity profile graph plotted against the pixel number. An on-axial row pixel which is going through the center of the x-ray source and along the direction of propagation is extracted using IDL programming and intensity grey-level versus pixel number graph plotted. Plotted graphs for two different shot numbers 470 and 482 are presented below in figure 4.10.
The length of the x-ray self-channel is estimated from the graph presented in the above figure 4.10. The estimated length of the channel is 1.37 mm, which is ~ 30 times longer than the length of Rayleigh-range (45 µm) of the focused pump-laser beam. The overall length of the x-ray source is 2.8 mm. It is very interesting to notice that some of x-ray source morphological types are producing self-guided channel (figure 4.9(c)) without entrance-zone. Intensity grey-level versus pixel number graph also plotted for those peculiar x-ray source morphological type and presented below in figure 4.11.
The above figure 4.11 shows that the interaction of pump-laser with Xe cluster molecules produces x-rays and the generated x-rays are trapped inside a self-guided dielectric waveguide and propagate in the direction of propagation until the defocusing mechanism diffract the self-channel radiation. The length of the self-channel in the particular peculiar case is 1.7 mm. It could be further observed that the channeled x-ray radiation inside the self-guided waveguide is having self-focusing and defocusing effects, which confirms the radiation trapped inside the self-channel is having coherent nature. The overall length of the x-ray source is 3 mm. The other very interesting propagation
dynamics observed is that the extension of self-channel waveguide after it diffract at the exit-zone. The observation of extension of self-channeling is presented below in the figures 4.12 and 4.13.

**Three times magnified Thomson scattering image**

![Graph](image)

**Plot of intensity profile versus pixel number**

![Graph](image)

Figure 4.12: Grey level versus pixel number graph for the on axis-pixel row going through center of the x-ray source and along the direction of propagation. The figure shows extension of self-channel after the diffraction point.

Overall-length of x-ray self-channel observed in the shot number 451 in between entrance and exit-zones is about 0.733 mm while overall length of the x-ray source is 2.3 mm. Length of extended-zone is 633 µm.
Length of x-ray channel observed in the shot number 387 is about 0.666 mm while overall length of the x-ray source is 2 mm. Extended length of self-channel after diffraction is about 400 µm. The 20 percent of experimental shots on August 17 2012 show the extended x-ray self-channel after the diffracted at the exit-zone. Particularly
some of the shots 400, 401, 410, 417, and 417 clearly reiterate the observation of extended exit-zone.

A simple diffraction theory is developed in the section 3.5.3 of chapter 3 for estimating diameter of the x-ray self-channels. The developed theory requires divergence-angle of the x-ray source at the end of the x-ray self-channel. For estimating the self-guided x-ray channel diameter, converging and diverging-angles of the x-ray source are estimated using IDL programming. Figures 4.14 and 4.15, presented below, briefly, are showing the analysis details.
The estimated convergence-angle is 16 degrees and the estimated divergence-angle is 26 degrees for shot number 372. The diffraction angle $\theta$ at the exit-zone is equal to 26 degrees. The triplet-aperture x-ray camera and TS diagnostics are not giving exact details about the wavelength of the x-ray source. However, the triplet-aperture x-ray camera filter details presented in the figure 4.2 is providing the range of wavelength of
the x-ray source. At this point, it is assumed that the center-wavelength of Xe x-ray source ($\lambda_c$) is 1.4 nm. Substituting the estimated values in the equation 3.54, the diameter of the x-ray channel is estimated. The estimated diameter of the x-ray channel for the shot number 372 is 1.14 times the center-wavelength of the Xe x-ray source.

The diameter of x-ray self-channel estimated for the shot number 480 where x-ray source is produced without entrance-zone. The detailed analysis is presented below in figure 4.15.
Estimated divergence angle at the exit-zone is 55 degrees, which is equal to the diffraction angle $\theta$. Substituting the estimated value for the $\theta$ and the center-wavelength of the x-ray source ($\lambda_x = 1.4$ nm) in the equation 3.54, the diameter of the x-ray self-
channel is estimated. The estimated radius of the x-ray self-channel for the shot number 480 is 0.61 times the center-wavelength of the x-ray source.

TS image presented in the above figure 4.15 shows multi-foci and de-foci points inside the dielectric waveguide where the propagating x-ray radiations are undergoing self-focusing and defocusing. The estimated x-ray energy from the aperture image for the shot number 480 is 30 µJ. Lower-bound value for the pulse-width of propagating x-ray should be less than or equal to pump-laser pulse-width. Therefore, the x-ray power (lower-limit value) trapped inside the self-channels is 85 MW. Further, the x-ray source morphology presented in the figure 4.4(c) (shot number 531) clearly shows bifurcation at the end of the x-ray source. The reference 76 is discussing a similar scenario takes place in UV pump-laser during its self-channel formation.

TS image data sets collected August 17 2012 show that self-focusing mechanism refocuses the x-ray radiation at the end of exit-zone. Twenty percent of experimental shots collected on August 17 2012 show the similar features. Converging-angle is estimated for shot number 410, which shows self-focusing beam at the end of exit-zone. The analysis presented below in figure 4.16 is similar to the previous analysis presented in the figures 4.14 and 4.15. Estimated convergence-angle from the analysis presented in figure 4.16 for the shot number 410. is 8 degrees.
The summary of the above analysis: The data sets collected by the triplet-aperture x-ray camera and TS show (i) the realized Xe M-shell x-ray sources are having convincing evidences for some degree of coherent nature and (ii) the realized x-ray source is also showing evidences for higher order of directionality.
The presented single-shot images captured by the calibrated single-shot triplet-aperture x-ray camera and the TS show that the wavelengths of the generated x-ray radiations are in the Xe \textit{M-shell}, and Xe \textit{L-shell} wavelength regimes. However, both diagnostics are not providing spectroscopic details of x-ray sources. Following section is discussion spectroscopic details of the realized Xe x-ray source.

### 4.2.3 Analyzing Single-shot Data Recorded by Calibrated Spectrometers from Xe X-ray Source

In this section, single-shot emission spectra captured by two von Hámos single-crystal spectrometer graphs, one in the axial-direction and other one in the transverse-direction are analyzed. The alignment details of the spectrometers are presented in the section 2.3.3.2.1 of chapter 2 and the calibration estimation are presented in the section 2.3.3.2.1.2 of chapter2. A sample raw data image and retrieved axial-emission spectrum from the average 31 pixel-rows of spectral image produced by the axial-von Hámos single-crystal spectrometers is presented below in figure 4.17.
Axial-von Ha’mos spectrometer spectrum

uv energy: 323 mJ
Xe temperature: 292 K
Xe pressure: 164 psig

Shot No.: 380
Exp. date: 08/17/2012

~200 µm

Axial-spectrum from average of 31 pixel rows

Figure 4.17: Axial-raw-data along with axial-Xe emission spectrum. In the spectrum, 31 pixel rows are averaged. The yellow lines show the extracted area and the red line goes through the maximum grey-level pixel row.
Xe emission spectra are analyzed in various methods using IDL programming. In the first-case, shown in the figure 4.17, fifteen pixel-rows above and below central maximum bright pixel-row is summed and plot against wavelengths. As shown in the figure 4.18 a pixel-row going through maximum-brightest pixel is extracted and plot against the wavelength. The presented figures 4.17 and 4.18 show the emission spectra in
the all two cases are almost similar. The von Hámos single-crystal spectrometers are an imaging device. Any wrong misalignment during the device fabrication and mechanical alignments of spectrometers will rotate the image-data and there is a risk of losing spectroscopic information when extract a single pixel-row. Therefore, in this thesis, the first case will be used as the emission spectrum of experiments.

Recorded spectra are compared with spectra recorded in one of the previous experiment presented in the references 28, 46, and 48. Even thou both (previous and current) experimental conditions are almost similar the spectroscopic features captured by the current spectrometers are showing additional new features in additions to some features, which are similar to spectroscopic features of a previous experiment captured by flat-crystal spectrometer. Sensitivity and dynamical range of diagnostics presented in this thesis is at least 100 fold greater than the diagnostics of previous experiments. Therefore, it is expected new features in the recorded spectra.

The sample Xe emission spectrum recorded by the flat-crystal spectrometer is presented below in figure 4.19 and the specifications of the flat-crystal spectrometer is presented in the section 2.3.3.2.1.2 of the chapter 2.
Where

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Ion</th>
<th>Transition</th>
</tr>
</thead>
<tbody>
<tr>
<td>p_{31}</td>
<td>Xe^{3+}</td>
<td>3d^4p \rightarrow 3d^5</td>
</tr>
<tr>
<td>p_{32}</td>
<td>Xe^{2+}</td>
<td>3d^4p \rightarrow 3d^5</td>
</tr>
<tr>
<td>p_{33}</td>
<td>Xe^{+}</td>
<td>3d^4p \rightarrow 3d^5</td>
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<tr>
<td>p_{34}</td>
<td>Xe^{0+}</td>
<td>3d^4p \rightarrow 3d^5</td>
</tr>
<tr>
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<td>Xe^{-+}</td>
<td>3p^6d^4p \rightarrow 3p^6d^3d</td>
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<td>Xe^{++}</td>
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<td>Xe^{+++}</td>
<td>3d^4s^6p^6f \rightarrow 3d^4s^5</td>
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<tr>
<td>s^4p_f^{24}</td>
<td>Xe^{++++}</td>
<td>3d^4s^6p^6f \rightarrow 3d^4s^5</td>
</tr>
<tr>
<td>s^6f^{25}</td>
<td>Xe^{+++++}</td>
<td>3d^4s^6f \rightarrow 3d^4s^5</td>
</tr>
<tr>
<td>S^2f^{26}</td>
<td>Xe^{++++++}</td>
<td>3d^4s^6f \rightarrow 3d^4s^5</td>
</tr>
</tbody>
</table>

The emission spectrum presented in the above figure 4.19 is compared with the emission spectrum data recorded in the experiments presented in thesis for performing calibration estimation as well as identifying the spectral-features. Figure 2.20, below, presenting axial and transverse-emission spectrum along with TS image for the shot number 639.
Figure 4.20: Axial and transverse-spectra collected by von Hámós single-crystal spectrometers along with TS image.
4.2.3.1 Peculiar Unidentified Spectral-feature at 1.34 nm

One of the immediate observations from spectra collected in the experiments show enhanced spectra-features at 1.34 nm wavelength, which was not observed in Xe emissions spectra collected by flat-crystal spectrometer. The observed feature of the enhanced radiation at 1.34 nm is more or less similar to features of x-ray amplifications at that wavelength. A literature research performed to get wind of any other research groups observed similar features. In 1912, C. T. R Wilson suggest that crystals with very distinct cleavage planes, such as mica, might possibly show strong specular (mirror-like) reflection of x-rays and it was experimentally verified by W. L. Bragg [77]. Research work presented in reference 78 also shows similar enhancement of x-ray radiation at about 0.67 nm in K absorption of Si single-crystal and amorphous SiO$_2$ single-crystal as presented below in figure 4.21.

![Figure 4.21](image)

**Figure 4.21:** K x-ray absorption spectra of (a) single-crystal Si (b) Amorphous SiO$_2$. The spectrum shows K-absorption edges of single-crystal Si and amorphous SiO$_2$ at 1840.3 eV (0.67 nm) and 1846.1 eV (0.67 nm) respectively.
In 1993, an experiment performed in XRIM also showed similar feature at 1.34 nm. In the experiment, BaF$_2$ target-material was irradiated with sub-picosecond UV and the spectrum were observed with von Hámos single-crystal spectrometer. The observed spectrum is presented in the reference 62 as unidentified feature at 13.40 ± 0.05 Å. Figure 2.22 below is presenting the spectrum.

![Graph showing spectrum](image)

Figure 4.22: Spectrum of BaF$_2$ irradiated with sub-picosecond UV radiation observed with a von Hámos single-crystal spectrometer on a single exposure shows unidentified feature at 1.34 nm [61].

Diffraction crystal of the von Hámos single-crystal spectrometer is Ruby mica (Muscovite; Phyllosilicate mineral of aluminum and potassium), which contains Si as one of its chemical components (KAl$_3$Si$_3$O$_{10}$ (OH)$_2$). As presented in the section 2.3.3.2 of chapter 2, the von Hámos single-crystal spectrometer is designed to operate at the first-order ($n = 1$). However, higher order diffracted x-rays are reaching the CCD image-plane and contribute to forms spectrum image. The wavelength ranges of von Hámos single-crystal spectrometer for different orders are presented below in table 4.1.
The table 4.1 shows that the Si and SiO₂ K-absorption edge wavelengths are falling within the second-order ($n = 2$) wavelength range (0.6-0.8 nm) of the von Hámó single-crystal spectrometer. As mentioned earlier, the Xe *M-shell* spectrum is superposition of Xe *M-shell* and Xe *L-shell* radiations. Therefore, first suspected reasons for the observation of enhanced x-ray radiation at 1.34 nm is that the 0.67 nm Xe x-ray radiations encounter resonant scattering at the K-edge of SiO₂ or Si elements of mica crystal and the second-order diffraction bring the 0.67 radiation to PI CCD ship and make it to overlap with 1.34 nm. In order to pinpoint exact wavelength of the observed feature and to determine whether the feature is coming from resonant scattering at the K-edge of SiO₂ or Si elements of mica crystal two diagnostics experiments are performed. Brief descriptions of the experiments are presented below in the following two paragraphs.

The first experimental approach involves with generation of another x-ray source from Kr clusters-laser-interaction in the wavelength range of 0.6 – 0.8 nm and experimentally tests the mica crystal efficiency at 0.67 nm due to the resonant scattering at the K-edge of SiO₂ or Si of mica crystal. If the second-order spectrum (where 1.34 nm is suppressed reasonably) captured by the von Hámó single-crystal spectrometer from the Kr x-ray source shows enhanced x-ray radiation at the wavelength 0.67 nm, one can determine whether the resonant scattering at the K-edge of SiO₂ or Si elements of mica

<table>
<thead>
<tr>
<th>Order ($n$)</th>
<th>Wavelength range (nm)</th>
<th>Wavelength range (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.2 – 1.6 (<em>Xe M-shell</em>)</td>
<td>1033 - 775</td>
</tr>
<tr>
<td>2</td>
<td>0.6 - 0.8 (<em>Kr L-shell</em>) (Si and SiO₂ K-absorption edge)</td>
<td>2067 – 1550</td>
</tr>
<tr>
<td>3</td>
<td>0.4 - 0.53</td>
<td>3100 - 2340</td>
</tr>
<tr>
<td>4</td>
<td>0.3 - 0.4</td>
<td>4133 - 3100</td>
</tr>
<tr>
<td>5</td>
<td>0.24 - 0.32 (<em>Xe L-shell</em>)</td>
<td>5167 - 3875</td>
</tr>
<tr>
<td>6</td>
<td>0.2 - 0.27</td>
<td>6200 - 4593</td>
</tr>
</tbody>
</table>

Table 4.1: Von Hámó single-crystal spectrometer different orders and corresponding wavelength range.
crystal enhance x-ray radiations at the 0.67 nm and the wavelength of the observed feature is at 0.67 nm (not 1.34 nm). Outcome of first experiment results is presented in section 4.3 of chapter 4, which confirm enhancement of x-ray radiation at the wavelength is 0.67 nm.

The second experimental study involve with introducing additional filters to the spectrometers for removing the lower-orders (first and second) diffracted x-ray radiations reaching the CCD chip image-plane. It means attenuating x-ray radiations shorter than 0.6 nm. A high-tech experimental setup is built for blinding the chip of axial-spectrometer with an additional 11.4 µm Ti filter during the experiment without breaking D chamber vacuum. The experimental setup is presented below in figure 4.23 along with filter transmission.

![Figure 4.23 (a)](image1.png)

11.4 µm Ti foil transmission graph for 0.2 - 1.6 nm

![Figure 4.23 (b)](image2.png)

Figure 4.23: (a) Experimental setup to introduce additional 11.4 µm filter foils to axial and transverse-spectrometers. (b) Filter transmission graph for 11.4 µm Ti foil. Wavelength rage for different orders of the von Hámos single crystal spectrometer are also shown in the figure (b).
The transmission graph clearly shows that introduction of the additional 11.5 µm Ti filter completely removes the first (1.2-1.6 nm) and second-order (0.6-0.8 nm) x-ray radiations reach the CCD chip. In the experiments, it was observed that the captured axial and transverse-Xe L-shell spectra are not showing the unidentified feature at corresponding 0.67 nm second-order wavelength. Therefore, it could be concluded, from the second experiment, that the unidentified peculiar feature at 0.67 nm (or 1.37 nm) is taking place only in the first and / or second-order mica diffraction. It means the wavelength of the observed feature should be in the wavelength range 0.6-1.6 nm.

One more experiment is performed to determine wavelength of the observed feature. Aluminum absorption K-edge is at the 0.74 nm ( 1.84 k eV) and transmission graph presented below in figure 4.24 shows the wavelength 0.67 nm corresponding to the unidentified peculiar feature line falls within the aluminum absorption K-edge well. Therefore, if the unidentified peculiar feature is at 0.67 nm, the features will be suppressed, with introduction of appropriate aluminum filter to the spectrometers. Transverse-von Hámos single-crystal spectrometer filter foil was replaced by a 2 µm Al filter foil while all other diagnostics and experimental parameters kept same. Collected experimental data reveal that the unidentified peculiar feature at 0.67 nm (or 1.37 nm) disappeared in the transverse spectra while almost all other features remained same. A sample spectrum is presented in figure 4.25. This experimental observation reconfirms that the wavelength range of the unidentified peculiar feature lies between 0.6–0.8 nm, which is corresponding to second order (n = 2) of von Hámos single-crystal spectrometer.
All possible allowed Xe atomic-transitions, which could produce x-rays at 0.67 nm, are carefully checked. It was found that there are no any allowed Xe atomic-transition, which could produce strong amplified x-rays at the 1.37 nm or 0.67 nm. Experimental observations are summarized below.

(i) BaF$_2$ experiment shows the peculiar unidentified amplified like feature at 1.34 nm. Spectrometer was operating in the first order ($n = 1$) (1.2 – 1.6).

(ii) Kr x-ray experiments shows the peculiar unidentified amplified like feature at 0.67 nm. Spectrometer was operating in the second order ($n = 2$) (0.6 - 0.8 nm).
(iii) Introducing 11.4 µm filter confirms that peculiar unidentified amplified feature is in the first and or second-order of the von Hámos single-crystal spectrometer (0.6-1.6 nm).

(iv) Replacement of 2 µm Al filter foil removed unidentified amplified like feature at 0.67 nm in the recorded spectra. Therefore, the wavelength of the feature is 0.67 nm.

(v) There is no allowed Xe atomic-transitions, which could produce amplified x-rays at 1.37 nm.

(vi) Xe x-ray emission recorded by flat-crystal spectrometer is not showing the peculiar unidentified amplified feature at 0.67 nm.

   Aluminum also one of the chemical elements of mica crystal and Al K-edge is at 0.79 nm, which is just outside the spectrometer range. If the resonant scattering at the K-edge of Si materials of mica crystal is producing the unknown feature at 0.67 or 1.34 nm, the Al also should produce an enhanced line at 0.79 nm in the second order and at 1.58 nm at the first order. This emission line was not experimentally observed, as these two lines are just outside to spectrometers range. Therefore, reconfiguration of the spectrometers to change its operating spectral range to cover the 0.79 nm (at second order) or 1.58 nm (at first order) and repeating the Xe experiment are necessary to observe the Al K-edge. If the resonant scattering of Al lines is observed, it could lead to conclude that the observed unknown feature at the 0.67 nm is due to the resonant scattering at the K-edge of Si materials of mica crystal. This experiment is one of future objective of this thesis.

*Although the above facts are convincing that the wavelength of peculiar unidentified feature is 0.67 nm and the observed line is due to the resonant scattering at the K-edge*
of Si materials of mica crystal, in order to come to a conclusion with 100% confident level it require further experiments, which is future objective of this thesis.

4.2.3.2 Xe Spectral-emission Features

Particular x-ray source morphological type (or experimental conditions) are producing Xe $M$-shell spectrum, which is similar to the spectrum recorded by the flat-crystal spectrometer (presented in the figure 4.19), is presented below in the figure 4.25 along with source image captured by the TS. A careful data analysis shows that not all of the spectra are similar to the spectrum presented below in the figure 4.25. Certain experimental conditions and x-ray source morphological types are producing spectra similar to the spectrum presented in the figure 4.25. Even thou, this thesis is not discussing the reason for various types of spectra it is also a future objective of this thesis.
Figure 4.25: Xe Emission transition lines are identified by comparing Xe emission spectrum recorded by axial- von Hámos single crystal spectrometer with Xe emission spectrum recorded by flat-crystal spectrometer (figure 4.19). The recorded spectrum shows additional spectral features, which was not observed in flat-crystal spectrometer. Transverse-spectrometer filter is 2 µm thick aluminum foil which completely attenuate Si K-edge feature in the transverse-spectra. The axial-spectrometer filter is 10 µm Be foil. During the experiment secondary N₂ gas tuned on. How an operation of DGSN alters x-ray, source energy is presented in the section 4.4 of chapter 4.
The Xe emission spectra of the experiment presented in this thesis are compared with the Xe emission spectrum recorded by flat-crystal spectrometer (presented in the figure 4.19) and the spectral features are identified. From the spectra presented in the figures 4.19 and 4.25 following observation are made with help of reference 46.

i. Formation of new 4f state with population. Xe electronic configuration is 1s² 2s²p⁶ 3s²p⁶d¹⁰ 4s²p⁶d¹⁰ 5s²p⁶ and 4f state is not exist in the ground state of neutral Xe atom.

ii. Xe M-shell spectrum shows one, two, and three 3d inner-shell vacancies and 4f → 3d and 4p → 3d transitions.

iii. The Xe M-shell spectrum is dominated by 4f → 3d transition arrays that occur in Xe ions at stages of ionization spanning from Xe²²⁺ to Xe³⁵⁺.

iv. In the transition 3d⁹4s²⁴p⁴f → 3d¹⁰4s²⁴p the 3d vacancy is coupled to the 4p outer electronic configuration which include an electron in the 4p state.

v. All of 4f → 3d transitions are coupled with 4s state with at least an electron in the 4s state.

It is very interesting to see abundance of 3d vacancies and population of 4f state. The intra-cluster model presented in the section 3.4.2.5 of chapter 3 explains observed 3d vacancies. However, it completely fail to explain the population of the 4f state, since no bound 4f electrons are present in the ground-state of neutral Xe atoms. The MPI-induced electron shake-up could promote an electron to the 4f level in an individual Xe atom. The process could result in the collateral formation of the 4d hole in the resulting Xe ion and subsequent 4f → 4d emission. The experimental observations presented in the reference 46 ruled-out the transition 4f → 4d. Therefore, there should be a different process, which
keeps the 4d sub-shell remains intact in the final state of the ion produced. As discussed in the section 3.4.2.6 of chapter 3, for atomic numbers \( Z > 58 \), the 4f state in neutral-atoms is collapsed to the inner-well forming an orbital, which is spatially localized at nearly the same radius as the orbital of the 4d state. Similarly, in many ionized systems for which \( Z < 58 \) can also possess collapsed 4f levels, since the attractive piece of the atomic potential is increased by the removal of outer-atomic electrons. Further, it is well established from photoionization studies involving multiple-electron ejection that the 4d, 5s, and 5p sub-shells exhibit substantial inter-shell coupling and behave in a collective fashion in a manner resembling a single super-shell, thereby causing the electron motions to involve a significant level of correlation [46]. Therefore, for satisfying all the experimental observations and predictions (from (i) to (v)), it appears that a combinations of MPI-induced orbital-collapse mechanism and the strongly-correlated electronic motions produce a special mechanism, which automatically forces the atom to be organized in the excited configuration and maximizing the radiation rate. The reference 46 indicates that the systematic of orbital-collapse phenomenon should occur for the 4f state in the region just below \( Z = 57 \) (I, Xe, Cs, Ba) and for the 5f state in corresponding atoms below \( Z = 90 \). The new systematic orbital-collapse mechanism is an excellent candidate for amplification in the multi-kilovolt range.

In x-ray laser field, when the axial and transverse-spectrometers are collecting simultaneously x-ray source spectral information from absolutely same x-ray source spatial location, recorded axial and transverse-spectra will show following two spectral properties if the x-ray source is behaving like a laser. The fundamental two spectroscopic evidences for laser-like sources are (i) amplified narrow spectral feature in the axial-
spectra, and (ii) observation of spectral-hole burning (missing radiation) in the transverse-exit-zone spectra. Recorded exit-zone-transverse Xe \textit{M-shell} spectra are not showing any prominent spectral hole-burning features. In the experiments presented in this thesis, the axial and transverse-von Hámos single-crystal spectrometer are not aligned to record spectral information from exactly same x-ray source spatial location. Therefore, direct comparison of axial-spectrum to transverse-spectra to determine laser-like properties might mislead experimentalist to wrong conclusion. However, signatures of the recorded axial and transverse-spectra are not exactly same. Both axial and transverse-spectra are showing different distinct features as shown below in figures 4.26, 4.27, 4.28, and 4.29.

Another interesting observation is that the exit-zone spectra are five-fold stronger than the entrance-zone spectra during the x-ray channel formation. Figures 4.26 and 4.27 show spectra along with TS image for no channel formation scenario and the figures 4.28 and 4.29 show the spectra for channel formation scenario.
Von Ha’mos axial and transverse-Xenon M/L-shell emission spectra, and Thomson scattering image

Figure 4.26: Transverse-entrance-spectrum is stronger than transverse-exit-spectrum when x-ray self-channel formation is not presence.
Von Ha’mos axial and transverse-Xenon M/L-shell emission spectra, and Thomson scattering image

Figure 4.27: Transverse-entrance-spectrum is stronger than transverse-exit-spectrum when x-ray self-channel formation is not presence.
Von Ha’mos axial and transverse-Xenon M/L-shell emission spectra, and Thomson scattering image

Figure 4.28: Transverse-entrance spectrum is stronger than transverse-exit spectrum when x-ray self-channel formation is presence.
Transverse-entrance spectrum is stronger than transverse-exit spectrum when x-ray self-channel formation is presence.

Figure 4.29: Transverse-entrance spectrum is stronger than transverse-exit spectrum when x-ray self-channel formation is presence.
Comparison of the figures 4.26 and 4.27 with figure the 4.28 and 4.29 clearly shows that the x-ray self-channel formations amplify the Xe M-shell x-rays. All the experimental data collected on August 17 2012 agrees with above discussion.

A few shots (less than 10 shots) show opposite observations where the entrance-spectrum is stronger than exit-spectrum while x-ray self-channel presence in between exit and entrance-zones. In those shots, the triplet-aperture camera images show presence of stronger x-rays in the entrance-zone than exit-zone. As presented in the section 2.3.3.2.2 of chapter 2 the transverse-von Hámos single-crystal spectrometer is collecting x-rays from entrance and exit-zone and forming corresponding spectra. Therefore, observed presence of stronger x-rays in the entrance-zone in the aperture-camera image is one of reasons for those shots to show stronger entrance-spectrum than exit-spectrum. However, more than 100 shots show relatively equal x-ray signal strength in entrance and exit-zone aperture-image while the exit-spectrum is stronger than entrance-spectrum and presence of x-ray self-channel.

### 4.2.4 Xe X-ray Source Brightness Estimation

This section is estimating brightness of Xe M-shell x-ray source. Brightness is defined as the number of x-ray photons per unit area per unit solid angle in unit time. In classical radiometry, the brightness (radiance) is defined by power \( P_{\text{x-ray}} \) radiated by unit surface \( dA \) into unit solid angle.

Definition of solid angle \( \Omega \) is presented below in equation 4.7.

\[
\Omega = \frac{\vec{n} \cdot d\hat{A}}{r^2} \tag{4.7}
\]
Where $\vec{n}$ is the unit-vector normal to the surface $d\vec{A}$, and $r$ is spherical surface radius.

In this thesis, Xe x-ray source brightness is estimated at a point where x-ray pencil beam hit and diffract on single-mica crystal. As a first step, incident x-ray energy on mica crystal is estimated using axial-von Hámos single-crystal spectrometer. Idea of x-ray energy estimation is similar to the x-ray energy estimation using triplet-aperture x-ray camera presented in the section 4.2.1.3 of chapter 4. During the diffraction of x-rays on the single-mica crystal x-ray energy is attenuate according to reflection-efficiency of mica crystal before it reach PI CCD chip. Therefore, in order to estimate incident x-ray energy on mica crystal, spectrometer efficiency is estimated as presented below.

Expression for efficiency ($B_{\lambda_x}$) for particular x-ray wavelength is presented below in equation 4.8 [44].

$$B_{\lambda_x} = \rho_m \nu_m \sin^2 \theta$$

(4.8)

Where $\rho_m$ is integrated reflectivity ($4.5 \times 10^{-5}$ radians (W·sr$^{-1}$·m$^{-2}$)) for 20 µm thick mica, 3$^{rd}$ order reflection), $\nu_m$ is acceptance angle in radiance (maximum allowed acceptance angle is 2.09 radians, the value is presented in the von Hámos single-crystal spectrometer design) and $\theta$ is the Bragg angle in degrees.

The acceptance angle ($\nu_m$) is estimated, as presented below in figures 4.30 and 4.31, for 2 different x-ray source morphological types. In the first case, the x-ray flux diverges in beginning of exit-zone and self-focusing mechanism focuses the x-ray flux at the end of exit-zone as shown below in the figure 4.30. Balance between self-focusing mechanism and diffraction mechanism decides divergence or convergence angle for x-ray pencil beam, which propagate, from exit-zone to mica crystal. It is experimentally
determined that the x-ray pencil beam reach the mica crystal with divergence angle of ~8 degrees. The details of experiments to estimate divergence-angle of x-ray pencil beam is not presented in this thesis. The first x-ray source morphological type presented in the figure 4.30 is observed in 80 percent of experimental data collected on August 17 2012.

In the second morphological type, presented in the figure 4.31, x-ray flux diverges in the exit-zone with 8 degrees divergence angle. Evidences for self-focusing effects were not observed in any of diagnostics and it is assumed the x-ray pencil beam is diverging with same divergence (8 degrees) angle until it reaches the mica-single crystal as shown in the figure 4.30.

Figure 4.30: TS image shows x-ray beam undergo divergence and then self-focusing mechanism focuses the beam towards on-axial direction (top-view). The von Hámos single-crystal spectrometer is introduced to TS image, which does not agree with Thomson scattering scale. Magnification of the TS is three.
The above two figures (4.30 and 4.31) and performed experimental x-ray beam propagation studies show that x-ray pencil beams are propagating with 8 degrees divergence-angles in all x-ray source morphological types. Therefore, all the necessary parameters, presented below, for estimating x-ray source brightness are equal for all x-ray source morphological types.

Radius \( r_{\text{source}} \) of x-ray beam at mica crystal is estimated using 8 degrees x-ray pencil beam divergence-angle and the distance between center of exit-zone and the mica crystal of von Hámos single-crystal (38 mm). Estimated x-ray beam radius \( r_{\text{source}} \) at a point where x-ray pencil beam hit mica crystal (shown in the figure 4.30) is 5.44 mm. The cross-sectional area of x-ray beam at that point is 93 mm\(^2\) and the estimated solid angle formed by the x-ray pencil beam at that point is equal to 64 milli-radiance.

Acceptance angle \( \psi_m \) of axial-spectrometer for the x-ray beam is estimated using radius of cylindrically bend mica crystal \( R_{\text{von}} = 26.7 \) mm and radius of x-ray beam
at a point where it first hit the mica crystal \((r_{source}=5.44\text{mm})\). The estimated acceptance angle \((\theta_m)\) is 23 degrees (0.41 radian). Spectrometers efficiencies \((B_{\lambda_{x}})\) are estimated using equation 4.2 and the estimated acceptance angle for different Xe \textit{M-shell} wavelengths (or Bragg angles). Estimated spectrometer efficiencies are plotted against Xe \textit{M-shell} wavelengths as presented below in figures 4.32 and 4.33.

X-ray energy and number of photons at certain Xe \textit{M-shell} x-ray wavelengths are estimated from the data image captured by the axial-spectrometer. The figures 4.32 and 4.33 below shows estimated x-ray energies in different Xe \textit{M-shell} wavelengths for the x-ray source morphological types presented in the figures 4.30 and 4.31 respectively. During the estimation of x-ray energy on CCD chip, x-ray energy attenuations factors such as spectrometer filter, propagation of x-ray beam through unionized Xe gas are not considered. However, the filter transmission, spectrometer efficiency for different Xe \textit{M-shell} x-ray wavelengths are incorporated into figures 4.32 and 4.33.

X-ray source energy estimation from triplet-aperture x-ray camera shows that \textit{L-shell} x-rays are contributing 7% to total x-ray energy produced by the interaction of pump-laser with Xe cluster molecules. The axial-energy estimation includes Xe \textit{M-shell} x-rays and Xe \textit{L-shell} x-rays. Therefore, 7% of total energy is subtracted from estimated total energy in order to estimate pure Xe \textit{M-shell} x-ray energy.
The figure gives axial and transverse-relative x-ray energy and Thomson scattering image. The plots show the relative x-ray energy in axial and transverse directions, with graphs for Xe M-shell x-ray energy at different wavelengths. The Thomson scattering image also includes a filter transmission graph and spectrometer efficiency graphs. Figure 4.32: The figure gives axial and transverse-relative Xe M-shell x-ray energy spectrum for x-ray source morphological type where x-ray source diverge at exit-zone and then in the self-focusing mechanism focuses propagating x-ray beam. X-ray attenuations factors are ignored in x-ray energy estimations and in the presented x-ray energy spectra. Filter transmission graph and the spectrometer efficiency graphs are also incorporated in the figure.
Figure 4.33: The figure shows axial and transverse-Xe M-shell x-ray energy spectrum for the x-ray source morphology where the x-ray source diverges at the exit-zone. Filter transmission graph and the spectrometer efficiency graphs are also incorporated into the figure. However, x-ray energy attenuations factors are ignored in the energy estimations and the x-ray energy spectra presented in the figure.
Systematic procedures for estimating number of incident photons on mica crystal at a certain wavelength $\lambda_x$ are presented below.

(i) Estimate energy at a particular wavelength $\lambda_x$ (shown in the figure 4.33)

(ii) Divide the estimated energy by spectrometer efficiency $B_{\lambda_x}$ and filter transmission, which gives x-ray energy with certain wavelength $\lambda_x$ incident on mica crystal.

(iii) Convert the x-ray energy and wavelength to the unit of eV.

(iv) Dividing x-ray energy with wavelength gives total number of x-ray photons with particular wavelength $\lambda_x$ incident on mica crystal.

All of above procedures are combined together to form a factor, which directly gives total number of x-ray photons at a certain x-ray wavelength incident on mica crystal when it multiplied by incident x-ray energy on the axial-spectrometer CCD chip at that wavelength. The factor is presented below by equation 4.9.

$$Fatcor = {\frac{1240 \times 6 \times 10^{12}}{\lambda_x \times B_{\lambda_x} \times T_{Fil}}}$$ (4.9)

Where $\lambda_x$ is x-ray wavelength, $T_{Fil}$ is filter transmission at wavelength $\lambda_x$ and $B_{\lambda_x}$ is efficiency of von Hámos single-crystal spectrometer at a wavelength.

As an example, total number of photons with the wavelength $\lambda_x = 1.34$ nm incident on mica crystal is estimated as follows. Estimated value for $B_{\lambda_x}$ from the figure 4.33 is $2.5 \times 10^{-5}$ radiance, and filter transmission is 0.41. Estimated value for the Fatcor is $5.4 \times 10^{20}$ and x-ray energy at 1.34 nm is $4.5 \times 10^{-8}$ µJ. Therefore, total number of photons at the wavelength $\lambda_x = 1.34$ nm incidents on mica crystal is $24 \times 10^{12}$. 
The pulse-width of the x-ray source is the final factor needed for estimating brightness of x-ray source. Theoretical works predicts that the pulse-width of x-rays, which is generated from hollow-atomic states, cannot be higher than pulse-width of pump-laser. Estimated pulse-width of pump-laser is in the order of sub-picoseconds (~350 fs). Therefore, the lower-limit of pulse-width of the x-ray source should be in the order of sub-picoseconds.

Total number of Xe M-shell photons reaching mica was estimated from the figure 4.32 by estimating number of photons in different wavelength and integrating them. Integrated total number of photons for the shot number 410 is 3.63 x 10^{17}. Area of the x-ray source at a distance 38 mm from center of exit-zone of the x-ray source is 93 mm^2 and solid angle at that point is 64 milli-radiance. Therefore, the integrated brightness of the x-ray source is in the order of 10^{26} photons / (sec. mrad^2.mm^2) with assumption, the pulse-width of the x-ray source is in the order of sub-picoseconds. The value for \( \Delta \lambda / \lambda_c \) (where \( \lambda_c \) is the center wavelength, which is equal to 1.4 nm) is 0.3. In the estimation, it is assumed that the beam is not attenuating during its propagation through plasma and Xe unionized gas column from center of source to the PI CCD chip. The total propagation distance (from center of exit zone to CCD chip) is over 6.5 cm, therefore the above mention assumption is not valid, and the brightness of x-ray source should be more than the estimated value. Further, theoretical works predicts that the pulse-width of the x-ray source is in attosecond regime. This thesis is not presenting any experimental measurement for estimating pulse-width of the x-ray source. However, if it is in attosecond regime the brightness will be in the order of 10^{32} photons / (sec. mrad^2.mm^2) which is comparable to the brightness of synchrotron radiations and the realized x-ray
source will be the brightest tabletop keV x-ray source of the 21\textsuperscript{st} century according to the tables 1.1, 1.2 and figure 1.3 of chapter 1. Further brightness of the developed Xe x-ray source is good enough for biological applications presented in the chapter 1.

Shot-by-shot x-ray energy flow within 16 degrees cone in the axial-direction is estimated with the help of above presented discussion and estimation of Xe x-ray source brightness. Estimated shot-by-shot x-ray energy flow within 16 degrees in the axial-direction is presented below in figure 4.34.

![Figure 4.34: Figure shows shot-by-shot x-ray energy flow within 16 degrees solid angle in the on-axial direction. The figure shows same features to the figure 4.7 where total x-ray source energy estimated from triplet-x-ray camera images](image-url)
Estimated average x-ray energy flow within 16 degrees cone in the on axial-direction is 43 µJ, which is 1 µJ higher than the total x-ray source energy estimated from triplet-aperture x-ray camera. Distance x-ray photons travel from center of x-ray source to the CCD chip of axial-spectrometer is almost equal to x-ray photons travel distance from center of x-ray source to the triplet-aperture x-ray camera filter (CCD chip is located behind the filter and the distance between CCD chip to filter and filter to x-ray source is same). Further, both diagnostics has same 10 µm thick Be filter foil and PI CCD chips are identical. Therefore, in both cases all x-ray energy attenuation-factors should be considerably equal for Xe \( M \)-shell x-rays. However, total x-ray energy estimated from triplet-aperture x-ray camera is smaller than the x-ray energy flow within 16 degrees solid angle in the on-axial direction. One of the possible explanation for this observation is x-ray self-channel mechanism, which channel x-ray to flow in the forward-direction and at the same time suppress in the transverse-direction. Therefore, one could conclude that the x-ray self-channeling mechanism compresses x-ray radiation into a dielectric waveguide and channel the x-ray radiations to flow in the on axial-forward direction.

In order to verify above explanation, forward and transverse-x-ray energy estimated from axial and transverse-spectrometers. Same pixel area (200 x 2048) extracted from axial and transverse-spectrometers CCD chip using ideal programming by keeping spectral image formed by the spectrometer at the center of the selected pixel-area. Xe \( M \)-shell x-ray energies are estimated using previously explained same techniques. Estimated shot-by-shot transverse and axial- Xe \( M \)-shell x-ray energies from 200 times 2048 pixels area are plotted and presented in figure 4.35 below.
The above figure 4.35 shows that Xe $M$-shell x-ray energy flow in the transverse-direction is at least three times smaller than the Xe $M$-shell x-ray energy flow in the forward-direction. This observation reiterate that the realized Xe x-ray source channel Xe $M$-shell x-ray radiation in the on-axial-forward direction.
4.3 X-ray Source Generation from Kr Hollow-atom States

This section discusses generation of Kr x-ray source from the interaction of pump-laser with Kr target-clusters. The experimental setup and diagnostics used for this experiment are exactly similar to the experimental setup and diagnostics of Xe M-shell x-ray source. Kr X-ray source image captured by the triplet-aperture x-ray camera is presented below in figure 4.36.

50 \(\mu\text{m}\) aperture x-ray camera image

![Image of x-ray source generated from Kr hollow-atom states and captured by triplet-aperture x-ray camera.](image)

- **uv energy**: 298 mJ
- **Kr temperature**: 291 K
- **Kr pressure**: 154 psig

**Shot No.**: 20  
**Exp. date**: 08/17/2012

Figure 4.36: Image of x-ray source generated from Kr hollow-atom states and captured by triplet-aperture x-ray camera.
The filter, which attenuate the 50 µm aperture for all the radiations longer than Kr \textit{L-shell} x-ray wavelengths is 10 µm Be foil. The filter transmission graph presented in the figure 4.36 shows that the x-ray produced from the hollow-atom states is in the Kr \textit{L-shell} kiloelectron volts regime. Analysis of triplet-aperture x-ray camera image is presented below in the following section.

4.3.1 Analyzing Single-shot Data Recorded By Triplet-aperture Kr X-ray Camera

The single-shot data collected by the triplet-aperture x-ray camera shows that the channel morphological structures of x-ray source produced from Kr hollow-atom states are similar to the previously observed channel morphological structures of Xe x-ray source produced from Xe hollow-atom states. Comparison of Xe \textit{M-shell} images and Kr \textit{L-shell} images reveal that the intensity levels of Kr \textit{L-shell} images are weaker than Xe \textit{M-shell} images for similar experimental conditions. X-ray energy of the Kr \textit{L-shell} x-rays generated from Kr \textit{L-shell} hollow-atom states are estimated in the following section and the estimated values are compared with the x-ray energy of the Xe \textit{M-shell} x-rays generated from Xe \textit{M-shell} hollow-atom states.

4.3.1.1 Energy Estimation of Kr \textit{L-Shell} X-rays

Energy of Kr \textit{L-shell} x-rays are estimated for every single-shot from triplet-aperture x-ray camera image. The details of estimation are similar to the estimation of Xe \textit{M-shell} x-ray energy and it is presented in the section 4.2.1.3 Figure 4.37, presented below, shows the result of the energy estimation.
The average energy of the Kr \textit{L-shell} x-rays at the triplet-aperture x-ray camera CCD chip is 18 \( \mu \text{J} \) which is almost two time smaller than Xe \textit{M-shell} x-rays (41 \( \mu \text{J} \)) produced from Xe hollow-atom states and 6 time bigger than the Xe \textit{L-shell} x-rays (3 \( \mu \text{J} \)). Attenuation factors, which attenuate the x-ray during their propagation from x-ray source to PI CCD chip, are not considered during the estimation. Therefore, actual x-ray source energy should be much higher than the 18 \( \mu \text{J} \) average energy. Filter
transmission graph for 2.65 mm thick Kr gas column shows (presented in figure 4.52) that the Kr \textit{L-shell} x-ray are attenuated by a reasonable amount.

4.3.2 Analyzing Single-shot Data Recorded by Thomson Scattering

The same TS optical setup, which was monitoring the Xe x-ray source, is now viewing the Kr x-ray source. The resolution of the TS is higher than the triplet-aperture camera. Therefore, TS setup perceived hidden information such as x-ray self-channel formation. In the following two sections TS is used as a tool to estimate x-ray self-channel length and diameter.

4.3.2.1 Estimating X-ray Self-channel Length

As in the Xe x-ray source the observation of channel formations was not clearly observed in the Kr x-ray source images captured by the TS diagnostics. In the Xe x-ray source data set, TS images showed that more than 90 \% of the experimental data are having x-ray self-channel formations. However, in the Kr source only 50 percent of experimental data shows the x-ray self-channel formation. One of the reasons for the disappearance of x-ray channel in 50 \% Kr x-ray source is that the diameter of Kr x-ray self-channels might be smaller than the Xe x-ray self-channels and the resolution of TS optical system might not be good enough to resolve x-ray self-channels in the Kr x-ray source. After presenting Kr x-ray self-channels and estimating the x-ray self-channel length, in the following two figures 4.38, and 4.39, and the diameter of x-ray channels are estimated.
The length of the x-ray self-channel for the shot number 92, presented in the figure 4.38, is 1 mm, which is 0.37 mm smaller than the Xe x-ray source presented in the figure 4.10 for shot number 470 and 0.7 mm smaller than Xe x-ray source presented in the figure 4.11 for shot number 482.
Length of the x-ray self-channel for the shot 159 presented in the figure 4.39 is 1.23 mm. These two x-ray self-channel length are almost smaller than the Xe x-ray self-channel length presented in the figures 4.12 and 4.13.
4.3.2.2 Estimating Kr X-ray Self-channel Diameter

A simple diffraction theory is developed in the section 3.5.3 of chapter 3 for estimating diameter of Kr x-ray self-channels and the section 4.2.2.1 is elaborating the idea of the estimation. As presented in the section 4.2.2.1, the developed theory requires divergence angle of the x-ray source at the exit-zone. For estimating the self-guided x-ray channel diameter, converging and diverging-angles of the x-ray source is estimated using code written in IDL software. Figures 4.40 and 4.41 presented below, briefly, is showing the analysis.
Figure 4.40: (a) TS raw data for shot number 319. (b) Shade-surf image shows the x-ray self-channel (c) Shows the extracted source-edge pixel points (d) Line-fit to the extracted source edge pixel points. The line-fit shows the source diverging nature.
Figure 4.41: (a) TS raw data for shot number 319. (b) Shade-surf image shows the X-ray self-channel (c) Shows the pixel locations at the edge of source (d) Linear fitting the source edge pixel locations (rows and columns).
The divergence-angle for the shot number 319 is 9 degrees and the divergence-angle for the shot number 240 is 7.5 degrees. The center wavelength of Kr x-ray source is 0.7 nm. Estimated divergence-angle and center wavelength are plugged in the equation 3.54 to estimate the diameter of the x-ray self-channel. Estimated self-channel diameter for the Kr x-ray source morphological type presented in the figure 4.40 is 2.2 nm. Estimated self-channel diameter for the x-ray source morphological type presented in the figure 4.41 is 3.83 nm. Comparison of estimated Kr x-ray self-channels diameters with Xe x-ray self-channels diameters show that Kr x-ray self-channels diameters are relatively equal or slightly higher than Xe x-ray self-channels diameters. In the estimations, it is assumed that the diffraction order is equal to one (n =1).

The entrance and exit-angle for Xe M-shell, Xe L-shell, and Kr L-shell are estimated from triplet-aperture x-ray camera images for different 5 single-shot, which has similar x-ray source morphological type and presented below in figure 4.42.
The figure 4.42 shows that the Xe $M$-shell sources, which has entrance and exit-zones are having smaller entrance-angle compared to the exit-angle. However, the Xe $L$-shell and Kr $L$-shell x-ray sources shows opposite effect, where entrance-angle is bigger than exit-angle.

Figure 4.42: Exit-angle versus entrance-angle graph for Xe $M$-shell, Xe $L$-shell and Kr $L$-shell for 5 different single-shot triplet-aperture x-ray camera images. The entire single-shot image has similar x-ray source morphological structures.
4.3.3 Analyzing Single-shot Data Recorded by Calibrated Spectrometers for Kr X-ray Source

A typical axial and transverse-spectra along with TS image is presented below in figure 4.43. The alignment of spectrometers and filter details are exactly similar to the alignment of spectrometers for Xe x-ray source.
von Ha'mos spectrometer axial and transverse-Krypton L-shell spectra and Thomson scattering image

Figure 4.43: Axial and transverse Kr emission spectra produced by the multi-photon excitation of Kr clusters with 248.6 nm pump-laser. TS image shows x-ray source.
Salient emission spectral signature lines captured by von Hámos single-crystal spectrometer graphs are compared with an emission spectrum presented in the reference 28 and below in figure 4.44 and corresponding transition for those spectral signature lines are identified.

4.3.3.1 Identifying and Explaining Kr Spectral-emission Features

The emission-spectral lines presented in the figure 4.43 and 4.44 show that in the wavelength range 0.6-0.77 nm the third Kr orbit (n=3) is coupled with the second Kr orbit (n=2). The strong lines observed in the 0.63–0.77 nm region uniformly involve transitions 3s, 3p, or 3d electrons. Further, the emission-spectral lines exhibit, in the 6.3-7.7 region, the multi-photon 3s → 2p transitions property consistently having a greater intensity at the wavelength 0.75 nm than the corresponding 3d → 2p lines. In order to
explain the observed spectral features the intra-cluster model presented in the section 3.4.2.5 of chapter two is recalled.

The 3s $\rightarrow$ 2p salient line at the 0.75 nm could be explained as follow. As presented in the intra-clusters model (presented in the figure 3.11) and MPI (presented in the figure 3.10) at the maximum intensity of $9 \times 10^{17}$ W/cm$^2$, 3s electrons cannot be removed by the tunneling process, although the full 3d shell is ionized for intensities greater than $2 \times 10^{18}$ W/cm$^2$. In addition to tunneling ionization, the collisional ionization cross-section for 3d state is approximately tenfold the value for the 3s state [28]. Therefore, retention of the 3s electron is again favored. It follows that in the region (0.6 nm – 0.8 nm) corresponding to L-shell excitation, particularly for intensities above $\sim 10^{18}$ W/cm$^2$, a higher probability exists for retention of 3s electrons than for 3d. This tendency should also be enhanced for the higher charge states seen, assuming a greater intensity for their production. Namely, the 3s/3d ratio should be greater for Kr$^{27+}$ than for Kr$^{26+}$, a feature that is reflected as shown in the above figure 4.44.

As presented in the reference 28 the 2p $\rightarrow$ 2s radiative decay is fast and electron collisional communication between the 2s and 2p states is slow, formation of Kr$^{26+}$ 2s2p$^6$3p levels by recombination from excited 2s2p$^6$Kr$^{27+}$ ions would have to be rapid. However, since the plasma conditions cannot support a recombination rate comparable to the radiative decay, it follows that the Kr$^{26+}$ excited states are directly produced in the excitation of the cluster. This explain the lines F and G involve 2s excitations. The same reasoning and conclusion hold in relation to the 3d $\rightarrow$ 2p satellite lines. Further, as presented in the section 4.2.3.1 of chapter 4, resonant scattering at the K-edge of Si
materials of mica crystal enhances spectral feature \( (3d \rightarrow 2p) \) at the wavelength 0.67 nm. Some further observations are systematically presented below in the following sections.

**4.3.3.2 Identifying Obvious Common Patterns from Single-shot Kr-Spectra Data Set**

Single-shot spectra captured by the axial and transverse-von Hámos single-crystal spectrometer and corresponding single-shot TS image along with all experimental parameters are processed to a single-page to view all the experimental parameters at a glance and to identify common features at various shots. Figures 4.45, 4.46, and 4.47 presented below, are showing processed spectra, TS image, and all experimental parameters for two different Kr x-ray source morphological types. Careful data analyses show that, axial-Kr spectral-line intensity at 0.75 nm for 3s \( \rightarrow 2p \) transition is always higher than the intensity of the spectral line at 0.67 nm for the 3d \( \rightarrow 2p \) transition. All the single-shots collected on August 17 2012 and the flat-crystal spectrum presented in the figure 4.44 agrees with this observation. However, the transverse-spectra show the opposite effect. In the 98 percent of the transverse-exit-zone spectra, collected on August 17 2012, spectral-line intensity at 0.75 nm for 3s \( \rightarrow 2p \) transition is smaller than the spectral-line intensity at 0.67 nm for the 3d \( \rightarrow 2p \) transition.

As observed in the Xe transverse-spectra, Kr transverse spectra also shows enhanced exit-zone spectral intensity compare to entrance-zone spectral intensity. The intensity enhancement nature during the propagation of x-rays from entrance-zone to exit-zone is similar to amplification effect. Out of total 367 shots, only seven shots were showing opposite nature. In the entire seven shots, the single-shot TS and triplet-aperture x-ray camera data show that presence of x-ray intensity in the entrance-zone is higher or
comparable to the exit-zone. Figure 4.47 is presenting one of the seventh shot. However, in these shots, which show high spectral intensity in the exit-zone compared to entrance-zone, the single-shot TS and triplet-aperture x-ray camera data show that presence of x-rays in the exit-zone is higher or comparable to entrance-zone (Figure 4.45, and 4.46).

Comparison of transverse-entrance spectra to transverse-exit spectra, are not showing evidences for spectral-hole-burning nature. However, carefully analyses shows the transverse-spectra are slightly different from the axial-spectra. Even-thou all primary emission-lines of both transverse and axial-spectra have similar features; the transvers-spectra are having more emission-lines those are missing in the axial-spectra.
Von Ha’mos spectrograph axial and transverse-Krypton L-shell spectra and Thomson scattering image

Figure 4.45: Axial and transverse-Kr spectra and TS image along with experimental parameters for shot number 77 shows that the spectral intensity of the entrance-spectrum is lower than the exit-spectrum. The TS image shows that x-ray intensity at the entrance-zone is comparable to the exit-zones-zone.
Figure 4.46: Axial and transverse Kr spectra and TS image along with experimental parameters for shot number 63. Spectral intensity of the exit-spectrum is higher than the exit-spectrum. The TS shows x-ray intensity is higher in the exit-zone compared to the entrance-zone. Further, it shows x-ray self-channel formation.
Von Ha’mos spectrograph axial and transverse-Krypton L-shell spectra and Thomson scattering image

Figure 4.47: Axial and transverse Kr spectra and TS image along with experimental parameters for shot number 308. Spectral intensity of the entrance-spectrum is higher than the exit-spectrum even though the TS image shows comparable x-ray intensity at the entrance and exit-zones.
The axial-von Hámos single-crystal spectrometer is moved along on axial direction of propagation of pump-laser beam and corresponding spectra are collected and compared. The analyses is presented in the following section.

4.3.3.3 Comparing Single-shot Kr Spectral Features at Various On-axial Locations

As presented in the section 2.3.3.2.1 of chapter 2, the axial-spectrometer is mounted on a five-axis positioner. The alignment of axial-spectrometer is presented in the figure 2.17. The axial-von Hámos single-crystal spectrometer initially aligned such a way the focal position of the spectrometer located at on-axial position B as shown in figure 4.48 ((X₁, 0, 0) according to the virtual Cartesian coordinate system defined in the section 2.3.3.21.1 of chapter). After collected enough data at this location, the spectrometer focal position is moved along the on-axial direction to new position D (X₁+2.5 mm, 0, 0). The transverse-von Hámos single-crystal spectrometer was intact during the experiments and IDL program extract the transverse-spectra from the spatial locations A, B, C, and D. Figure 4.48 below is comparing transverse-spectra at various on-axial positions along with the axial-spectrum collected from spatial location D.
Von Ha’mos spectrographs spectra at different on-axial-focal locations and Thomson scattering image

Figure 4.8: Axial- Kr spectrum is recorded from point D and transverse-spectra extracted from on-axial-spatial locations at point B, C, and D using IDL programming. The transverse-spectra extracted from positions B, C, and D show that spectral intensity drastically drops from point B to point D. In the other hand, the axial-spectral intensity remains relatively same at the points B and D.
In general, in the field of x-ray lasers, there are three fundamental spectroscopic evidences are necessary to confirm lasing action. They are

(i) The lasing line should be high compared to non-lasing lines.

(ii) Lasing lines should dominate in spectrum.

(iii) The *saturated amplification* regime where the signification part of the population inversion should be converted into the lasing signal line.

The figure 4.48 shows that the intensity of the transverse-spectrum is decreasing drastically from point B to D. The transverse-spectrum intensity at point D is nearly zero while the axial-spectrum intensity is nearly the same strength as it was at the point B. This observation is a convincing evidence that 3d \( \rightarrow \) 2p and 3s \( \rightarrow \) 2p lines are enhanced during propagating in the direction of propagation of the pump-laser beam. Further, the figure 4.48 clearly shows that spectral peaks of the spectra collected from point C are slightly shifted to the right when it compare to the spectral peaks of spectra collected from points A, and B. This observation implies that the Kr spectrum experience a red-shift when x-ray beam propagates in the on-axial forward direction.

### 4.3.4 Estimating Brightness of Kr(L) X-ray Source

The methods of Kr x-ray source-brightness estimations are similar to the performed Xe x-ray source brightness presented in the section 4.2.4 of chapter 4. Figure 4.49, presented below, is showing distribution of x-ray energy in Kr *L-shell* wavelength regime.
During the experiment, the axial-spectrometer moved along the direction of pump-laser and single-shot data are recorded. Data analyses show that the Kr x-ray attenuation factors, which attenuate x-rays during its propagation, is ignored in the energy estimation. The filter transmission graph and spectrometer efficiency graph in the Kr L-shell wavelength regime are incorporated in the figure.

**Figure 4.49:** Axial and transverse Kr x-ray energy spectra along with TS image. The factors, which attenuate x-rays during its propagation, is ignored in the energy estimation. The filter transmission graph and spectrometer efficiency graph in the Kr L-shell wavelength regime are incorporated in the figure.
sources are having average 5 degrees divergence-angle. The details of the analyses are not presented in this thesis. However, the divergence-angle estimation from the TS image (shown in the figure 4.39, 4.40) agrees with the estimation.

As shown in the figure 4.29, the Kr x-rays hit the mica crystal at a distance of 38 mm from center of Kr x-ray source. Kr x-ray source diameter is estimated, from raw-axial-spectrometer image, at a spatial location where focus of axial-von Hámos single-crystal spectrometer is initially located (point B in the figure 4.47). Estimated Kr x-ray source size at this location is 300 µm and estimated Kr x-ray source diameter at a distance of 38 mm from center of x-ray source is 7 mm. Estimated effective acceptance-angle (\(\nu\)) of spectrometer is 0.260 radiance. Estimated solid angle at this position is 26 milli-radiances. Estimated number of photons and Kr x-ray source in the shot number 16 are presented below.

Total number of Kr \textit{L-shell} x-ray photons at a distance 38 mm from center of exit-zone is in the order of \(10^{17}\).

\textit{Total number of photons at the wavelength 0.67 nm at a distance 38 mm from center of exit-zone is in the order of \(10^{16}\).}

\textit{Total number of photons at the wavelength 0.75 nm at a distance 38 mm from center of exit-zone is in the order of \(10^{16}\).}

A pulse can be defined as a transient in a constant background. The shape of this pulse is the shape of this transient. Since half-maximum quantities are experimentally easier to measure, the relationship between the duration and spectral bandwidth of the laser pulse can be written as [79]

\[\Delta\nu\Delta t = K\] (4.10)
where $\Delta \nu$ is the frequency bandwidth measured at full-width at half-maximum (FWHM), $\Delta t$ is the FWHM in time of the pulse and $K$ is a number which depends only on the pulse shape. In the most cases, $K$ number is always smaller than one.

One can also calculate the minimum time duration of a pulse giving a spectrum with $\Delta \lambda$, (nm) at FWHM, central wavelength $\lambda_c$, (nm) and the speed of light (m/s) $c$.

$$\Delta t \geq K \frac{\lambda_c^2}{c \Delta \lambda}$$  \hspace{1cm} (4.11)

Estimated value of $\Delta \lambda$ for 0.67 nm is $35 \times 10^{-4}$ nm.

Estimated **pulse-width** for 0.67 nm is $43 \times 10^{-17}$ seconds.

The value $\Delta \lambda_{0.67}/\lambda_{0.67}$ is $53.73 \times 10^{-4}$.

Estimated value of $\Delta \lambda$ for 0.75 nm is $41 \times 10^{-4}$ nm.

Estimated **pulse-width** for 0.75 nm is $45 \times 10^{-17}$ seconds.

The value $\Delta \lambda_{0.75}/\lambda_{0.75}$ is $53.73 \times 10^{-4}$.

*Estimated brightness of Kr x-ray source at 0.67 nm is in the order of $10^{28}$ photons $s^{-1} mm^{-2} mrad^{-2}$.*

*Estimated brightness of Kr x-ray source at 0.75 nm is in the order of $10^{28}$ photons $s^{-1} mm^{-2} mrad^{-2}$.*

It is assumes in the above estimations that the divergence angle of the Kr x-ray beam from the transitions 3d $\rightarrow$ 2p ($\sim$ 0.67 nm) and 3s $\rightarrow$ 2p (0.75 nm) lines are having same divergence-angle of main x-ray source. However, it is expected the divergence-angles of the beam is smaller than the assumed divergence-angle, which will increase brightness in reasonable amount. Shot by shot axial-energy is estimated and plot against shot number and presented below in figure 4.50.
The average axial-Kr \textit{L-shell} x-ray energy in the forward direction within 8.5 degrees of divergence-angle is 8\,\mu\text{J}, which is almost half of total Kr \textit{L-shell} x-ray energy (18\,\mu\text{J}) estimated from triplet-aperture x-ray camera analyses. The comparison of axial-x-ray energy and total energy implies that the generated x-ray is channeled in the forward axial-direction. Shot by shot axial and transverse x-ray energy in the 200 x 2048 CCD...
pixel area of axial and transverse-spectrometer CCD chip is estimated and presented below in figure 4.51.

The above figure 4.51 shows that ratio of axial-x-ray energy to the transverse x-ray energy flow is ~1.7 and confirms that x-ray self-channel mechanism channels x-ray radiations in the forward-direction.
4.4 Optimizing Realized X-ray Sources

Optimizing the realize x-ray sources is one of primary objective of the research work presented in this thesis. X-ray source optimizing parameters presented in the chapter 2 is summarized below.

(i) Brightness of pump-laser, and focus spot spatial-pointing-stability on target-clusters: Provide optimized pump power at controllable fashion to ionized target-clusters. Presented experimental results confirm that the new laboratory, and laser-system infrastructure designs and constructions optimized brightness of pump-laser.

(ii) Target-clusters density and size: Optimize quality of target-medium. Different types of sonic nozzles are experimentally tested to find a suitable nozzle, which optimize the density and size of target-clusters. A commercial nozzle double gas sonic nozzle (DGSN) is redesigned to optimize target-clusters density, and to reduce x-ray attenuations during the propagation of x-rays through target-medium. This section is discussing experimental results of the DGSN.

(iii) Experimental diagnostics: A tool to understand physics behind the x-ray sources. Presented results in the chapter 4 show the dynamical range and resolution of calibrated diagnostics are improved at least 100 times compare to the previous diagnostics.

(iv) Analyzing experimental results: Understand physics behind the x-ray sources open new windows to optimize x-ray source.
In addition to above four x-ray source-optimizing parameters, spatial positions of pump-laser focal-spot to the target-cluster and DGSN poppet opening timing play important roles in optimizing brightness of x-ray sources. During the experiments, DGSN is being moved to different spatial locations until real-time measurements shows optimize x-ray source. Figure 4.52 is presenting typical spatial location of pump-laser focus spot relative to DGSN orifice. Further, the DGSN poppet opening timing varied, during experiments, in the order of microseconds until the real time measurements show optimized x-ray source. In typical experiments, DGSN opens around 780 ns prior the Hurricane laser emits a seeding laser pulse. Beside the second x-ray source optimizing parameters (density and size of target-clusters) all the other x-ray source optimizing parameters are discussed previously. Discussions of dependency of x-ray source brightness on target-gas temperature and DGSN operation are presented below.

It is experimentally shown that the cluster formation is strongly augmented by cooling the flow in the jet. The experimental results presented in the reference 42 clearly shows that the cooling the target-gas increases x-ray intensity. The experimental results are not presented in this thesis. However, it is presented in the reference 42.

In addition to lowering the target-gas temperature, the hybrid DGSN is designed to optimize target-clusters density and to reduce the x-ray attenuation during the propagation of x-rays through target-medium. The designs and operations of DGSN are presented in the section 2.3.2 of chapter 2. Figure 4.52 presented below shows the alignment of UV pump-laser focus spot to target-clusters in a typical experimental setup, which produces optimize x-ray source. The pump-laser focus spot is aligned to the entrance central orifice edge of sonic nozzle. Once the x-ray is produce from interaction
of pump-laser with target-clusters, the generated x-rays are propagating at least 2.65 mm through target-gas medium, which will attenuate propagating x-rays, before it diffract single-mica crystal of axial-spectrometer. Transmission graph for 2.65 mm Xe and Kr, presented below in the figure 4.52, gives an idea of attenuation during the propagation of x-rays through 2.65 mm Xe, Kr gas column.

Figure 4.52: Alignment of focal-spot to DGSN orifice in a typical experimental setup. Figure shows gas divergence boundary when DGSN operation is turned OFF.
The density of Xe gas in the above transmission graph is $0.6 \times 10^{-2}$ g/cm$^3$ and the density of Xe gas in the above transmission graph is $0.4 \times 10^{-2}$ g/cm$^3$. Density measurements performed in various nozzle and presented in the reference 41 shows that the optimized density produced by all types of sonic-nozzles are in the order of $10^{19}$ molecules/cm$^3$. The estimated density value for Xe and Kr densities from density measurement is comparable to the presented Xe and Kr density in the above figure 4.53.

As discussed in the section 2.3.2 of chapter 2, the DGSN is designed for increasing Xe cluster density and for reducing Xe gas column through which produced Xe $M$-shell x-rays are propagating. Designed DGSN compresses Xe target-gas into a smaller volume with definite boundaries as shown in the figure 2.53. DGSN interferogram results presented in the section 2.3.2 of chapter 2 confirms the success of
the design. Experiment performed on August 12 2011, N gas turned ON at the shot number 80. The figure 4.53 (a) and (b), presented below, compare target-gas area before and after N gas turned ON. In the figure 4.54 Thomson scattering image shows the area where target Xe gas molecules are presence and figure 4.53, further, clearly shows that the N gas compresses the Xe gas into small volume.
Figure 4.5: Overlay of DGSN to triplet-aperture x-ray camera and Thomson scattering image (a) when N gas is turned OFF and (b) when N gas turned ON. The figure shows that the secondary N2 gas confines the primary Xe gas into a smaller volume shown in the dotted lines. Blue circles shows primary-gas orifice and red circles shows secondary gas orifice.
Xe x-ray source Xe *M-shell* x-ray energy in the axial-direction is estimated from the axial-spectrometer data after and before N gas turned ON. Both data are presented below in figure 4.55.

**Plot of Axial-Xe(M) x-ray energy versus shot number**

- Average energy: 250 mJ
- Xe pressure: 179 psig
- Xe temperature: 239 K
- Exp. date: 08/17/2012

![Graph showing x-ray energy vs shot number](image)

Figure 4.55: Axial-Xe *M-shell* x-ray energies before and after N gas turned ON. At shot number 80, N gas was turned ON. Average energy without DGSN action is 21 µJ and average energy with the operation of DGSN is 41 µJ.

The figure 4.55 clearly shows that the operation of DGSN doubles average axial-x-ray energy. The data set presented in the above figure 4.54 further shows that DGSN operation, in some of the shots, boost axial-x-ray energy almost five times. During the experiment, Prometheus preamplifier malfunctions, in some of shots, result to deliver
very low pump-energy to target-clusters. In this thesis, the shot at which Prometheus malfunction taken place is termed as “bad shot”. In the figure 4.55, red box shows “bad shots”. If one ignores the “bad shots” the average axial-x-ray energy during the operation of DGSN will be considerable higher than the presented 41 µJ axial-x-ray energy.

The axial and transverse Xe $M$-shell x-ray energy is estimated by extracting 400 x 2048 CCD pixel from transverse and axial-spectrometers. Results are presented below in figure 4.56.

**Plot of transverse versus axial-Xe(M) x-ray energy**

![Graph showing comparison of transverse and axial-energy with and without operation of DGSN.](image)

Figure 4.56: Comparison of transverse and axial-energy with and without operation of DGSN.
The figure 4.56, presented above, shows that the operation of DGSN support x-ray self-channel formation and increases x-ray energy flow in the forward axial-direction. X-ray source morphological image captured by the triplet x-ray camera and Thomson scattering, presented in the figure 4.54 (b), confirms the observations of x-ray self-channel formation during the operation of DGSN.

Various experiments performed on DGSN shows that optimization of axial-x-ray energy depends on following parameters.

I. Primary and secondary gas stagnation pressures.

II. Actuating time delay between primary and secondary solenoid-poppet of sonic nozzles.

III. Lowering both primary and secondary gases temperature.

IV. Density and size of clusters.

V. Laminar flow pattern of primary and secondary gases.

Experimental evidences presented above shows that operation of DGSN is a promising candidate to boost axial-x-ray energy. Further, it shows that the operation of DGSN increases axial-energy few order higher than previously used any other techniques to increase axial-x-ray energy. Therefore, a new upgraded, DGSN is designed with help of Polish academic of science who is pioneer in DGSN designs. New designed DGSN has very fine tune-ability to control stagnation pressures of primary and secondary gases separately, and to control primary and secondary gases spraying delay time separately. Further, the DGSN consist Peltier cooling techniques, which enables primary and secondary gases to lower their temperatures with higher controllable fashion to operate DGSN at desired lower temperature. The orifice of DGSN is machined such a way to
produce smooth surface to make laminar gas flows. Preliminary experiments performed to study the gas flow pattern, cluster formation, and temperature controllability gives clear evidences for its reliable operation and laminar gas flow. The new DGSN is being introduced to experimental setup. A complete study of DGSN operations and axial x-ray energy will be performed in near future and it is the last future objective of this thesis.

😊------------------- END OF CHAPTER 4 -------------------😊
5. CONCLUSION

A decade of research works performed to develop and study the x-ray sources presented in this thesis. On June 13 2009, first experiment performed in newly built system. Since then the system is constantly recording single-shot data file. Each recorded file includes (1) 248 nm pump-laser energy (mJ), (2) target gas sonic-nozzle stagnation pressure (psig), (3) target gas temperature (K), (4) triple x-ray source images captured by triplet-aperture x-ray camera, (5) a transversely observed spectrum, (6) an axially observed spectrum, (7) transversely captured Thomson scattering image, (8) seed beam focal-spot image, and (9) sonic-nozzle orifice image captured by an aperture x-ray camera. Total amount of recorded data is over 3 terabytes and total number of single-shots files is over two hundred thousands. This thesis is presenting only few important key findings of a decade research work to develop and study the Xe and Kr x-ray sources and this section, conclusion, conclude the experimental finding presented in this thesis.

Remodeled laboratory and laser system is producing 2.5 µm seed-beam focused spot waist-size with less than 2.5 µm beam pointing stability. The estimated final amplified pump-laser focused spot diameter meter is 10 µm, which is producing $5 \times 10^{17}$ W/cm$^2$ intensity level. Developed novel design and pump-laser beam alignment procedure drastically reduced time needed for pump-laser beam and diagnostics alignment and reproduces the alignments precision enough to perform day-to-day experiments in similar fashion.

A hybrid Double Gas Sonic Nozzle (DGSN) is designed to produce target gas cluster at densities of solid targets. Previously sonic-nozzles used in XRIM experiments
were capable of spraying only single-target-gas to produce target clusters. The designed DGSN is spraying target-gas at the center and the low-Z secondary-gas in the primary gas annular region to increase cluster density and to reduce x-ray attenuation during the propagation of x-ray through unionized target gases. Estimation of total number of target-gas molecules available inside the Rayleigh range for excitation and the estimated power of pump-laser shows that 1 W/atom pumping power is achieved.

X-ray source images captured by the triplet-aperture x-ray camera and the transmission graph of filter foils to attenuate all radiations longer than desired radiations confirms that the x-ray sources are in the keV x-ray regime. There are various different x-ray source morphological types are observed during the experiments. Although more experiments needed to understand completely the reasons for the shot-by-shot variations of x-ray source morphological types, we learned how to select desired type of morphological type by tuning experimental conditions. Comparison of simultaneously captured Xe *M-shell* x-ray radiations and Xe *L-shell* x-ray radiations clearly shows that the radiation volume and morphological type of Xe *M-shell* and Xe *L-shell* are identical and equal. This observation concludes that Xe *L-shell* radiations are caused by Xe *M-shell* radiations like a chain reaction scenario.

Thomson scattering diagnostic data shows that generated x-ray radiations are trapped and channeled by a dielectric waveguide whose diameter is in the order of x-ray source center wavelength. Multi self-focusing and defocusing points are observed inside self-channels. The minimum power trapped inside the channel is in the order of MW. Length of x-ray self-channels are varies shot by shot. The minimum length of observed self-channel is at least 30 times longer than the Rayleigh range of pump-laser focal-spot
on target-clusters. Divergence and convergence-angles of the x-ray sources are estimated with help of TS image and aperture camera images. The data sets shows in Xe **M-shell** x-ray sources entrance-angle is smaller than exit-angle but in the Xe **L-shell** and Kr **L-shell** x-ray sources entrance-angle is bigger than exit-angle. Commonly observed features of Xe **M-shell** and Kr **L-shell** x-ray sources are presented below in tabular format.

<table>
<thead>
<tr>
<th>X-ray source type</th>
<th>Xe <strong>M-shell</strong> x-ray source</th>
<th>Kr <strong>L-shell</strong> x-ray source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength range (nm)</td>
<td>1.2 – 1.6</td>
<td>0.6 – 0.8</td>
</tr>
<tr>
<td>Minimum brightness (Photons s⁻¹ mm⁻² mrad⁻²).</td>
<td>10²⁶ (with assumption of sub picosecond pulse-width)</td>
<td>10²⁸</td>
</tr>
<tr>
<td>Average x-ray energy at a distance 3.5 cm (in the forward direction) from center of source (µJ).</td>
<td>43</td>
<td>8</td>
</tr>
<tr>
<td>Average length of x-ray self-channel (mm).</td>
<td>1.2</td>
<td>1.1</td>
</tr>
<tr>
<td>Average x-ray source diffraction angle at the end of self-channel.</td>
<td>5</td>
<td>4.5</td>
</tr>
<tr>
<td>Repetition rate (Hz)</td>
<td>0.1</td>
<td>0.1</td>
</tr>
<tr>
<td>Enhance spectral feature wavelength (nm).</td>
<td>1.34</td>
<td>0.67, 0.75</td>
</tr>
<tr>
<td>Pulse-width at enhanced spectral feature</td>
<td>Not available</td>
<td>450 attosecond</td>
</tr>
</tbody>
</table>

Table 5.1: Characterized x-ray sources parameters.

In the Xe axial and transverse-spectra, an enhanced spectral feature is observed at 1.34 nm. Although the feature is similar to amplifications, a study of this feature shows that this feature is arising due to the properties of mica crystal of von Hámos single-crystal spectrometer where x-ray diffract before reach CCD chip. Spectroscopy studies shows that during the x-ray self-channel formations the transverse spectra collected from entrance-zone of self-channeled waveguide is weaker than the transverse-spectra collected from exit-zone of x-ray self-channel. In the Kr x-ray sources, transverse-spectra and axial-spectra are collected simultaneously from various on-axial locations. Recorded data sets shows that the strength of Kr axial-spectral features, particularly 3d \( \rightarrow \) 2p transition at 0.67 nm and 3s \( \rightarrow \) 2p transition at 0.75 nm are increasing in the on-axial direction of
propagation of pump-laser while corresponding transverse-spectra shows that the strength of those particular transitions are fainting in the on-axial-direction of propagation of pump-laser. Recorded spectra show in the both x-ray sources hollow-atomic states are produced during the interaction of pump-laser with target-cluster and transitions from outer-shells are producing ultrabright x-rays.

Almost in all experiments performed in the newly built system, the DGSN is used as a single-gas sonic-nozzle similar to previously used sonic-nozzles, which spray only single target-gas for target-cluster formation. Experiments performed to test the design of DGSN. In the experiments, primary target gas sprayed at the center and the secondary low-Z gas sprayed at different pulse timing and different stagnation pressure in the target-gas annular area for increasing cluster density while reducing x-ray attenuation during the propagation of x-rays through unionized target-gas column. Experimental results presented at the end of chapter 4 clearly show that the operation of DGSN increases average Xe M-shell axial x-ray energy at least a factor of 2. This experimental observation confirms that DGSN actions optimize axial x-ray energy significantly higher than any other previously experimented efforts in XRIM such as lowering target-gas temperature to increase x-ray energy. Observed optimum axial x-ray energy at a distance 3.5 cm from center of Xe x-ray source is 100 µJ. The action of DGSN, in addition to boost axial x-ray energy, suppress x-ray energy flow in the transverse-direction and hence support the x-ray self-channeling mechanism.

Realized ultrabright x-ray sources shows convincing evidences for laser-like natures such as coherent and directionality. Further research works are needed to confirm the observations. The achieved brightness at the keV wavelength regime makes the
ultrabright keV x-ray source as unique x-ray sources and such x-ray source will be a identical tool to lithographical applications, high-precision holographic micro fabrication, non-destructive testing with phase-sensitive projection imaging. Finally this thesis concludes that the developed x-ray sources from Xe and Kr hollow atom-states are the brightest available laboratory-scale keV x-ray source to date.

😊--------------------- END OF CHAPTER 5 ---------------------😊


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APPENDICES

Appendix 1.1: Needed spatial-resolution at different wavelength.

![Diagram A.1.1: Spatial resolution versus wavelength.](image)

Appendix 2.1: Hurricane laser system with tripler-unit

![Diagram A.2.1: Schematic diagram of Hurricane laser system and tripling unit.](image)
Appendix 2.2: Schematic diagram of fourth generation laser system.

Diagram A.2.2: Schematic diagram of the fourth generation laser system.
Appendix A.2.3: D chamber technical drawing
Appendix A.2.4: Closed looped water system for LLG 50 PRO

Figure A.2.1: Closed-looped water circulating system of LLG 50 PRO
Appendix 2.5: Schematic diagram of XRIM triggering system

Diagram A.2.4: Schematic diagram of triggering system.
Appendix 2.6: Quantum-efficiency of front illuminated PI camera.

Figure A.2.2: Quantum efficiency graph of PI front illuminated CCD camera.

Appendix 2.7: Transmission graph of interference filter.

Figure A.2.3: Thomson scattering interference filter transmission graph.
Appendix 2.8: Effective focal length shift of Thomson scattering micro-spot focusing-objective for different wavelengths.

Figure A.2.4: Effective focal length shift of Thomson scattering micro-spot focusing-objective for different wavelengths.
Appendix 3.1: ionization potential and charge states of Xe

<table>
<thead>
<tr>
<th>Charge state Z</th>
<th>State</th>
<th>Ionization potential (i_p) (eV)</th>
<th>Wave length (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1s(^-)</td>
<td>12.1</td>
<td>102.5</td>
</tr>
<tr>
<td>2</td>
<td>2s(^-)</td>
<td>21.1</td>
<td>58.77</td>
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<td>3</td>
<td>3s(^-)</td>
<td>23.2</td>
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<td>6</td>
<td>6s(^-)</td>
<td>71.8</td>
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<td>7</td>
<td>7s(^-)</td>
<td>92.1</td>
<td>13.46</td>
</tr>
<tr>
<td>8</td>
<td>8s(^-)</td>
<td>106.0</td>
<td>11.70</td>
</tr>
<tr>
<td>9</td>
<td>9p(^-)</td>
<td>171.0</td>
<td>7.25</td>
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<td>10</td>
<td>10p(^-)</td>
<td>202.0</td>
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<td>358.0</td>
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<td>857.0</td>
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<td>26</td>
<td>26p(^-)</td>
<td>1495.0</td>
<td>0.83</td>
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<table>
<thead>
<tr>
<th>Charge state, Z</th>
<th>State</th>
<th>Ionization potential (i_p) (eV)</th>
<th>Wave length (nm)</th>
</tr>
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<td>3d(^+)</td>
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<td>0.83</td>
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<td>28</td>
<td>3d(^+)</td>
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<td>1877.0</td>
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<td>3d(^+)</td>
<td>1987.0</td>
<td>0.62</td>
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<td>33</td>
<td>3d(^+)</td>
<td>2085.0</td>
<td>0.59</td>
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<td>3d(^+)</td>
<td>2211.0</td>
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<td>3d(^+)</td>
<td>2302.0</td>
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<td>3p(^+)</td>
<td>2639.0</td>
<td>0.47</td>
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<td>3p(^+)</td>
<td>2728.0</td>
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<td>3p(^+)</td>
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<td>3p(^+)</td>
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<td>3p(^+)</td>
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<td>0.38</td>
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<td>3p(^+)</td>
<td>3334.0</td>
<td>0.37</td>
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<td>44</td>
<td>2p(^+)</td>
<td>7663.0</td>
<td>0.16</td>
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<tr>
<td>45</td>
<td>2p(^+)</td>
<td>7893.0</td>
<td>0.16</td>
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<td>46</td>
<td>2p(^+)</td>
<td>8143.0</td>
<td>0.15</td>
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<td>47</td>
<td>2p(^+)</td>
<td>8381.0</td>
<td>0.15</td>
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<tr>
<td>48</td>
<td>2p(^+)</td>
<td>8987.0</td>
<td>0.14</td>
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<td>49</td>
<td>2p(^+)</td>
<td>9257.0</td>
<td>0.13</td>
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<td>50</td>
<td>2s(^+)</td>
<td>9582.0</td>
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<tr>
<td>51</td>
<td>2s(^+)</td>
<td>9813.0</td>
<td>0.13</td>
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<td>52</td>
<td>1s(^+)</td>
<td>40425.0</td>
<td>0.03</td>
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<tr>
<td>53</td>
<td>1s(+)</td>
<td>41211.0</td>
<td>0.03</td>
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</tbody>
</table>

wave-length range: \(\lambda \approx \frac{1240}{E(eV)}\) nm

Table A.3.1: Ionization potential and charge states of Xe.

**Tentative wavelength range for Xe M-shell and Xe L-shell**

Xe **M-shell** wavelength range: \(~0.95 – 1.6\) nm (1300 – 775 eV)

Xe **L-shell** wavelength range: \(~0.24 – 0.33\) nm (5.2 – 3.8 keV)

\[ E(eV) \approx \frac{1240}{\lambda(nm)} \]
Appendix 3.2: Xe absorption edge

<table>
<thead>
<tr>
<th>Xe absorption edge</th>
<th>keV</th>
<th>Å</th>
</tr>
</thead>
<tbody>
<tr>
<td>K</td>
<td>34.56</td>
<td>0.36</td>
</tr>
<tr>
<td>L-I</td>
<td>5.45</td>
<td>2.27</td>
</tr>
<tr>
<td>L-II</td>
<td>5.10</td>
<td>2.43</td>
</tr>
<tr>
<td>L-III</td>
<td>4.78</td>
<td>2.59</td>
</tr>
<tr>
<td>M1</td>
<td>1.14</td>
<td>10.83</td>
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</table>

Table A.3.2: Xe absorption edges.

Appendix 3.3: Kr absorption edge

<table>
<thead>
<tr>
<th>Kr absorption edge</th>
<th>keV</th>
<th>Å</th>
</tr>
</thead>
<tbody>
<tr>
<td>K</td>
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<td>0.87</td>
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<tr>
<td>L-I</td>
<td>1.92</td>
<td>6.45</td>
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<tr>
<td>L-II</td>
<td>1.72</td>
<td>7.18</td>
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<td>L-III</td>
<td>1.67</td>
<td>7.40</td>
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</table>

Table A.3.3: Kr absorption edges.

Appendix 3.4: Intensity and values for Xe OTBI and Keldysh parameter $\gamma$ for Xe charge states.

<table>
<thead>
<tr>
<th>$l_0$</th>
<th>Z</th>
<th>OTBI</th>
<th>State</th>
<th>$\gamma$</th>
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<tbody>
<tr>
<td>12.1</td>
<td>0</td>
<td>None</td>
<td>5p$^5$</td>
<td>None</td>
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<tr>
<td>21.1</td>
<td>1</td>
<td>7.9E+14</td>
<td>5p$^3$</td>
<td>7.39</td>
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<tr>
<td>32.1</td>
<td>2</td>
<td>1.1E+15</td>
<td>5p$^4$</td>
<td>5.99</td>
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<tr>
<td>46.7</td>
<td>3</td>
<td>2.1E+15</td>
<td>5p$^3$</td>
<td>4.97</td>
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<tr>
<td>59.7</td>
<td>4</td>
<td>3.2E+15</td>
<td>5p$^3$</td>
<td>4.39</td>
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<tr>
<td>71.8</td>
<td>5</td>
<td>4.3E+15</td>
<td>5p</td>
<td>4.01</td>
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<tr>
<td>92.1</td>
<td>6</td>
<td>8.0E+15</td>
<td>5s$^2$</td>
<td>3.54</td>
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<tr>
<td>2106</td>
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<td>9</td>
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<td>4d$^3$</td>
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<td>421</td>
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<td>1.60</td>
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<tr>
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<td>4p$^3$</td>
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<tr>
<td>583</td>
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<td>1.3E+18</td>
<td>4p$^4$</td>
<td>1.34</td>
</tr>
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</tbody>
</table>

Table A.3.4: Intensity and values for Xe OTBI and Keldysh parameter $\gamma$ for Xe charge states.
VITA

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Won prestigious Arunasalam Memorial Prize for the best performance in the undergraduate Physics final exams, 1999.

PROFESSIONAL MEMBERSHIP: Member of Optical Society of America, 2003 – Present.
Founder and public relation officer of Sri Lankan Graduate Student Association of UIC, 2003 – Present.

PUBLICATIONS: “Comparative analysis of stable ultrahigh power-density 248 nm channels formed in Kr and Xe cluster targets,” Alex B. Borisov, John C. McCorkindale, Sankar Poopalasingam, James W. Longworth, and Charles K. Rhodes, submitted to J. Phys. B.

“Demonstration of Kr(L) amplification at $\lambda = 7.5$ Å from Kr clusters in self-trapped plasma channels,” Alex B. Borisov, John C. McCorkindale, Sankar Poopalasingam, James W. Longworth, and Charles K. Rhodes, submitted to J. Phys. B.


“Observation of a curve crossing mechanism in the field ionization of inner-shell excited single \( \text{Xe}^{33+} (2\,\ell\,\ell) \) and double \( \text{Xe}^{34+} (2\,\ell\,\ell) \) vacancy states,” Alex Boris Borisov, Shahab Firasat Khan, Ervin Rácz, Sankar Poopalasingam, John Charters McCorkindale, Ji Zhao, Joel Bernard Fontanarosa, Yang Dai, James William Longworth, and Charles K. Rhodes, IEEE, 48: 806-813, 2012.

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“Spatially resolved observation of the spectral hole burning in the Xe(L) amplifier on single \( (2\,\ell\,\ell) \) and double \( (2\,\ell\,\ell) \) vacancy \( 3d \rightarrow 2p \) transitions in the \( 2.62 \,\text{Å} < \lambda < 2.94 \,\text{Å} \) range,” Borisov Alex B., Racz Ervin, Khan Shahab F., Sankar Poopalasingam, McCorkindale John C., Zhao Ji, Fontanarosa Joel, Dai Yang, Boguta John, Longworth James W, and Charles K. Rhodes, J. Phys. B., 43: 045402, 2010.


“Single-pulse characteristics of the Xe(L) amplifier on the Xe\(^{35+}\) \( (3d\rightarrow2p) \) transition array at \( \lambda \approx 2.86 \,\text{Å} \),” Borisov Alex B., Song Xiangyang, Zhang Ping, McCorkindale John C., Khan Shahab F., DeJonghe Richard, Sankar Poopalasingam, Zhao Ji, Boyer Keith,

😊 ———— No end for research ———— 😊