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DEPARTMENT OF ENERGY CONTRACT  
DE-FG03-92-SF19200

AN EXPERIMENT TO DEMONSTRATE A NITROGEN RECOMBINATION X-RAY  
AMPLIFIER USING HIGH-DENSITY PLANAR GAS JET LASER TARGET

March 31, 1996

Prepared by

J. G. Pronko and D. Kohler

Advanced Technology Center  
Lockheed Martin Missiles and Space Company  
Palo Alto, California, 94304-1191

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Sponsored by the U. S. Department of Energy for work conducted for the National  
Laser Users Facility, Laboratory for Laser Energetics, University of Rochester.

**ABSTRACT**

The results of an experiment to search for lasing in atomic transitions at x-ray energies in  $N_2$  gas target plasmas using ultra-short laser pulses is presented. Particular emphasis was placed on a search for a predicted 24.7 nm optical-field-ionization (OFI) induced lasing line from the Li-like nitrogen ( $N^{4+}$ ,  $3d \rightarrow 2p$ ) transition. The excitation laser was a multi-terawatt Cr:LiSrAlF<sub>6</sub> laser system operating at a wavelength of 825 nm and a pulse duration of 135 fs located at the Lawrence Livermore National Laboratory. Experimental conditions were optimized and a series of Li-like (including the 24.7 nm  $N^{4+}$   $3d \rightarrow 2p$ ) lines were observed and identified. Further experimental studies are required before an attempt at measurement of any potential lasing gain can be made.

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## Section 1

## INTRODUCTION

At the time of the proposal, recombination x-ray lasing at 18.2 nm had already been demonstrated by irradiating solid carbon targets [1,2]. The scheme uses a high-power laser to create a fully stripped carbon plasma which radiatively cools, while magnetically confined, until the plasma conditions are such that dielectronic recombination to the H-like state (C VI) dominates the various rate processes. This process preferentially populates the higher Rydberg levels which then cascade to lower levels by radiative decay. As the electrons decay to lower levels, population inversions can arise because the radiative rates out of a hydrogenic state increase as the energy of the level decreases allowing the lower level of a given pair to empty more rapidly. In the case of carbon, population inversion occurs between the  $n = 3, 4$  and the  $n = 2$  levels with lasing action observed for the  $n = 3$  to  $n = 2$  transition at 18.2 nm which was measured gain coefficient of  $\sim 5 \text{ cm}^{-1}$ .

In order to achieve a shorter wavelength, which scales as  $Z^2$  for hydrogenic ions, elements above C must be used. Because several relatively benign and cheap low-Z elements are gases, a gas-jet laser-target capable of producing a planar sheet of gas and which can be irradiated by a line-focussed laser beam suggests itself. Such a jet possess several advantages over solid targets, not least of which is the convenient access to the lower-Z plasmas of N, O, and Ne. In addition, C and F are available in gaseous forms as well. Thus, an x-ray laser based on these low-Z elements represents the lowest-energy approach to a potential series of coherent, high-brightness x-ray beams which can then be used by a broader scientific community.

The high-density gas-jet target used in this experiment has been developed under a Lockheed Martin Advanced Technology Center internally funded independent x-ray

laser research program. The objective of this program is to explore the viability of using a laser heated gas jet target to generate a relatively uniform plasma column suitable as an x-ray laser medium. Lockheed Martin has worked closely with the Plasma Research Corporation Inc.<sup>3</sup> to design and fabricate the nozzles and high-pressure valves which compose the gas jet target system. One potential advantage of such a gas target is that it should be possible to make a planar gas flow which should be initially quite uniform and which has density gradients much less severe than those found in laser-produced plasmas from solid targets.

The proposed experiments were to have been performed on DOE's Glass Development Laser (GDL) at the Laboratory for Laser Energetics (LLE), University of Rochester, which operates at a wavelength of 1054-nm. From the standpoint of coupling the laser radiation to the jet it is desired to use the longest pulse available, preferably on the order of 1-ns. The beam was to be line focussed to a 1-cm length or as close to this length as possible so that the entire 1-cm wide sheet of gas was illuminated. The line-focus was to be at a point approximately 2-mm above the nozzle exit orifice where atomic densities as high as  $10^{19}$  atoms/cm<sup>3</sup> could be achieved which implies electron densities of a few times  $10^{20}$  cm<sup>-3</sup>.

The contract for this investigation commenced on 2 January 1992 with P. C. Filbert as Principal Investigator. Preparations were made for the experiment to be performed on the GDL laser. Diagnostic equipment were prepared and calibrated, and the gas jet was optimized for use on this facility. The experimenters were later informed that the availability of the laser would be delayed due to the lack of funding for the upgrade and operation of the GDL facility. Eventually, the facility became permanently unavailable and was closed. Sometime after this period of delays, P. C. Filbert left the company and was no longer available to act as Principal Investigator. J. G. Pronko and D. Kohler assumed the responsibility for the contract at which time only 65% of

the original funding was available after all of the early preparations prior to and during the 3 years of delays.

It became apparent that it was necessary to find an alternate facility at which to perform the experiment. The Lockheed Martin R&D Division in Palo Alto, Ca is conveniently located near Lawrence Livermore National Laboratory (LLNL). Since the commute time is approximately 45 to 60 minutes between the two facilities and a close relationship has been maintained in the past with the laser groups at LLNL, it was decided to form a collaborative effort with one of the groups for this experiment. Carrying out the research at LLNL has the additional advantage of substantially reducing travel expenses and technical delays would not have as great an impact on the program resources.

Discussions were held with M. D. Perry of LLNL's Short-Pulse Laser and Diffractive Optics group at the LLNL. The group maintains multi-terawatt Cr:LiSrAlF<sub>6</sub> laser systems which operate at 825 nm at femtosecond (fs) pulse durations. The investigation of the lasing schemes outlined in the original proposal required much longer pulse durations. However, there have been recent suggestions of other lasing schemes in nitrogen which could be studied using the fs multi-terawatt lasers. One of these processes is described below.

This alternate approach to the achievement of a recombination x-ray laser became available with the recent advent of fs terawatt (TW) laser systems. This approach makes use of optical-field-ionization (OFI) of the lasant atom. In the OFI ionization approach [4] a very high intensity ( $\geq 10^{16}$  W/cm<sup>2</sup>) short pulse ( $\leq 100$  ps FWHM) is directed into the lasant gas or plasma. For an appropriate set of conditions, choice of lasant atom, laser intensity and wavelength, and gas or plasma density the atom can be ionized down to a hydrogen-like or lithium-like species, or even be completely ionized. This ionization occurs through tunneling of the electron from the given atom and is due to the very high imposed optical electric field of the laser pulse and occurs within a very few

optical cycles. This complete and very sudden ionization then forms the starting point for the development of a transient inversion during the subsequent recombination.

The recombination process itself can be very rapid and efficient since the OFI ionization process characteristically results in a relatively cold electron plasma which rapidly fills the higher Rydberg states prior to the development of the recombination cascade. The first part of the pulse which ionizes the higher states produces the coldest part of the resultant electron energy distribution and it is essential to select operating parameters to avoid any subsequent substantial heating of the electron spectrum (especially the coldest part). Detailed analyses of the approach for a number of possible lasants including lithium-like-neon and lithium-like-nitrogen (the latter being the subject of the experiment described in this report) have been carried out indicating the parameters required for lasing can be made consistent and should be achievable.

Amendt et al<sup>4</sup> propose a demonstration experiment of OFI plasma x-ray lasing at 24.7 nm in nitrogen. They outline the predicted conditions for which x-ray lasing in this Li-like nitrogen ( $N^{4+}$ ,  $3d \rightarrow 2p$ ) transition could take place. The initial goal of this experiment was to study and optimize the experimental conditions required for producing lasing in this transition using nitrogen gas targets and ultra short laser pulses. The ultimate goal, after optimization of the experimental parameters, was to measure the lasing gain with a planar gas jet target as the lasing medium. The latter involves striking the gas jet on the planar surface with a line focused beam while comparing the measured x-ray transition yield along the line focus and perpendicular to the line focus.

Permission was received from the DOE to perform the experiment at LLNL with the intent of exploring the concept of OFI lasing schemes using nitrogen gas jet targets.

## Section 2

## EXPERIMENTAL APPROACH

## Laser Facility.

The experiment was performed using a multi-terawatt Cr:LiSrAlF<sub>6</sub> (Cr:LiSAF) laser system maintained by the Short-Pulse Laser and Diffractive Optics group at the LLNL. It is a flash-lamp-pumped system capable of producing fs pulses exhibiting peak powers greater than 5 TW at a wavelength of 825 nm. Chirped pulse amplification in a Cr:LiSAF regenerative amplifier produces 15-mJ pulses up to a 5 Hz repetition rate. Further amplification in Cr:LiSAF yields recompressed pulse energies of 1 J and a pulse duration of less than 135 fs up to a 1.0 Hz repetition rate.

The system begins with a self-mode-locked Ti:Sapphire oscillator that produces transform-limited 110-fs pulses of 8 nJ at 76 MHz. This pulse train is collimated with an  $f = 1.6$  m lens and is injected into a diffraction-grating pulse stretcher. The stretcher uses 1800-line/mm gold-coated holograph diffraction gratings and two 60-cm focal-length cemented achromatic doublet lenses corrected for spherical and chromatic aberration at 825 nm. Four passes through the stretcher produce enough positive group-velocity dispersion to stretch the pulse from 110 fs to 500 ps.

At this point the p-polarized pulses are injected into a ring regenerative amplifier cavity. The regenerative amplifier utilizes a single 4 mm x 50 mm flash-lamp-pumped 1.5% doped Cr:LiSAF rod as the amplifying medium. A single pulse is selected for amplification by application of half-wave voltage (5800 V) to a potassium dihydrogen phosphate (KDP) Pockels cell placed inside the cavity.

The system cavity dumps the amplified pulse with the Pockels cell after the desired number of round trips in the cavity. The Pockels cell is triggered at the peak of the single pass gain in the LiSAF amplifier, which closely follows the flash-lamp pulse. The output at this stage is approximately 15 mJ. Final amplifiers are capable of taking the pulse to a 1J level.

Some of the data collected during the experiment consisted of using a prepulse to form the initial ionization. This was accomplished by splitting out 25% of the beam and delaying the main pulse by 14 ns.

#### Gas Jet Configurations.

Two types of gas jets were used in this experiment. These were the high-density axially-symmetric gas jet and the high-density planar gas jet. Each of these are described below.

High-Density Axially-Symmetric Gas Jet. This gas jet is based on a solenoid driven valve mated to an axially-symmetric supersonic DeLaval nozzle with a nominal Mach number of approximately 8. The throat and exit diameter of this ceramic nozzle are 150  $\mu\text{m}$  and 1.5 mm, respectively. The valve assembly is relatively light and small and is in the shape of a cylinder 3.8 cm long with a diameter of 1.8 cm. The gas reservoir is connected to a cylinder of pressurized gas and a small volume of gas is released each time the poppet valve is withdrawn from the nozzle entrance opening by the high-speed solenoid carriage. The volume of gas depends on the gas reservoir pressure and the duration of time the poppet valve is held away from the entrance to the nozzle. The solenoid is driven by a high-voltage square wave pulse and the poppet is returned to the closed position by spring pressure. The length of the driving pulse determines the temporal length of the gas-jet plume. The gas jet was designed under

Lockheed Martin Advanced Technology Center funding and built by Plasma Research Corporation<sup>3</sup>.

The atomic density produced by this circular nozzle was measured by a nuclear scattering technique with 2-MeV  $\alpha$ -particles elastically scattered by the atoms in the jet. This measurement is described in a recent publication<sup>5</sup>.

High-Density Planar Gas Jet. The high-density planar gas jet target was developed under Lockheed Martin Advanced Technology Center funding. The Plasma Research Corporation has been a subcontractor in this effort and has translated the functional target requirements into completed hardware.

The valve mechanism uses a relatively new magneto-strictive material (Terfenol<sup>TM</sup>) as the active element. Upon applying a magnetic field to the material it expands and actuates a lever arm which pulls a linear plunger from the nozzle entrance allowing the high-pressure gas in the plenum to escape through the nozzle. The nozzle is designed for a Mach 4 flow and has a one-dimensional DeLaval cross section with a 1-cm long and 0.1 cm wide exit aperture. With this design the valve which can be pressurized to 3000 psi, opened in roughly 100  $\mu$ s, remain open for 200 - 300  $\mu$ s, and closed in 100  $\mu$ s. This is a significant improvement over high-pressure valves which use a solenoid as the valve actuating mechanism because these have opening and closing times in the millisecond regime. It is desired to keep the valve open as short a time as possible in order to keep the gas loading of the vacuum chamber as low as possible. The expected density performance of the planar gas jet target is based on the actual measured performance of the gas-jet target having a circular exit orifice described above.

### X-ray Spectrograph Configuration.

The spectrograph was housed in a 15.24 cm diameter vacuum pipe mounted on the straight through port of the target chamber. The x-rays entered the spectrograph at a grazing incident angle of  $3^{\circ}$  onto a 5 cm long curved grating with 1200 lines/mm. The x-rays scattered by the grating were subsequently reflected by a carbon mirror into the camera recording instrumentation. The positions of thin film filters, the grating, and carbon mirror and the angle of incidence were such that the straight through path of the laser beam to the recording instrumentation was occluded. Experiments were performed with the recording instrumentation being either a CCD camera (1000 x 200 pixels) or a streak camera. The spectrograph was set up to record x-ray lines from approximately 10 to 50 nm. In addition to experiments performed with nitrogen gas, data was occasionally collected with argon gas. The latter data was used for reference and energy calibration purposes.

### Streak Camera Experiments.

The streak camera was a Kentech Instruments low magnification re-entrant x-ray streak camera. The camera is optimized for the ultra sensitive recording of x-ray images and spectra from laser produced plasmas. The low (approximately x 1.2) magnification allows a 25 mm length cathode (P11 or P20) to be used within a 40 mm diameter intensifier window. The camera can operate at 6 sweep speeds between 1 and 80 mm/ns (i.e. 1000 to 12.5 ps/mm, respectively). The cathode for the present experiments consisted of 100 nm Al. The camera was used in conjunction with the x-ray spectrograph.

**Video Camera Configuration.**

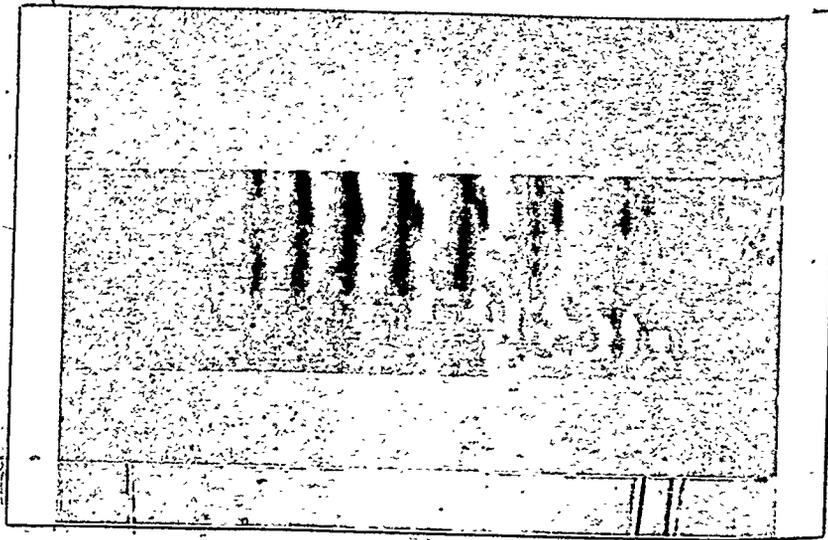
As part of the procedure to optimize conditions for the planar gas jet target, a COHU solid state video camera was set up to view the visible light from the laser generated plasma. It was anticipated that this experiment would provide information on the dimensions of the ionized portion of the planar gas jet. A lucite window was placed on a port on the side of the chamber and the camera was coupled to a computer for capturing the video frames. The jet was mounted on the chamber port opposite the video camera. The gas jet fired in the direction of the camera and an alignment was performed to be sure the gas jet intersected the laser beam at the optimum position. Video frames were taken as a function of gas jet backing pressures between 100 and 600 psi and laser beam energies between 100 and 300 mJ.

## Section 3

**EXPERIMENTAL RESULTS**

The experiment was performed at LLNL during the two week period between 27 March and 7 April 1995. Five days of laser time were dedicated to the experiment in each of those weeks. J. G. Pronko participated in the experiment during both weeks while D. A. Kohler could only participate during the 1st week. The experiment was performed in collaboration with T. Ditmire (a graduate student of M. D. Perry from U.C. Davis) and T. D. Donnelley (a graduate student of R. W. Falcone from U.C. Berkeley).

The initial experiment performed consisted of collecting data with the x-ray spectrograph and CCD camera described in the previous section. The spectrograph was set up to collect an x-ray spectrum over the region of 10 to 50 nm. Data was collected for both N<sub>2</sub> and Ar using the planar gas jet. Since previous experimental data was available using an Ar gas target, the intent was to use the spectral data collected with the Ar gas target as a source for reference and calibration of the experimental configurations. The gas jet was aligned with the laser beam hitting the gas jet plane edge on. The laser was fired in a single pulse mode at total beam energies between 20 and 250 mJ with back pressures between 400 and 600 psig. A search of parameters was performed to optimize the position of the planar gas target and the timing between the arrival of the laser pulse and the firing of the gas jet. Typical data collected under these conditions are illustrated in Figs. 1 and 2 for Ar and N<sub>2</sub> gas respectively. The upper portion of each of the figures presents a photograph of the spectrum while the lower portion of each figure presents a scan of the spectrum along the energy axis. The spectra are typical of harmonic generation of the laser beam. Harmonic generation is influenced by both the state of ionization and the laser beam polarization. As ionization builds up



FILE "N03281B.SPE" Horizontal line-out  
ROWS #53 TO 147 Y FROM 1.24800 TO 3.50400  $\Delta Y$ (EDGES)=2.28000

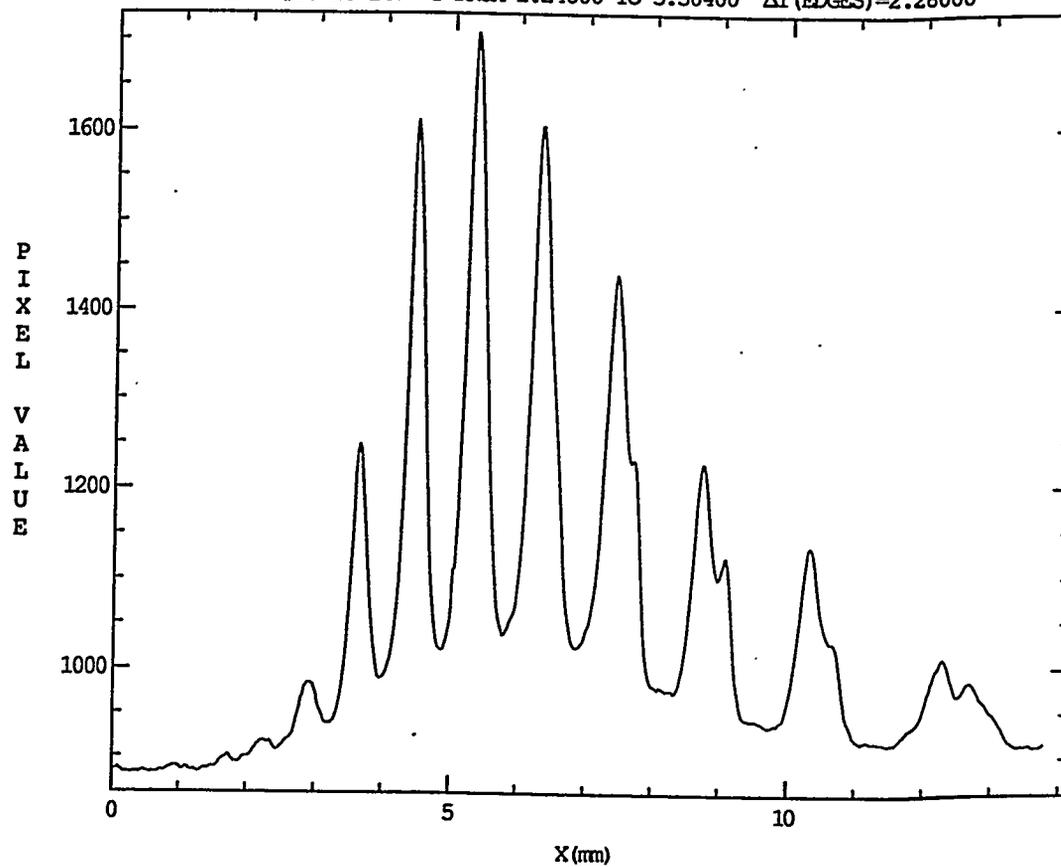


Fig. 1 The x-ray spectrum generated with  $N_2$  gas and the planar gas jet.

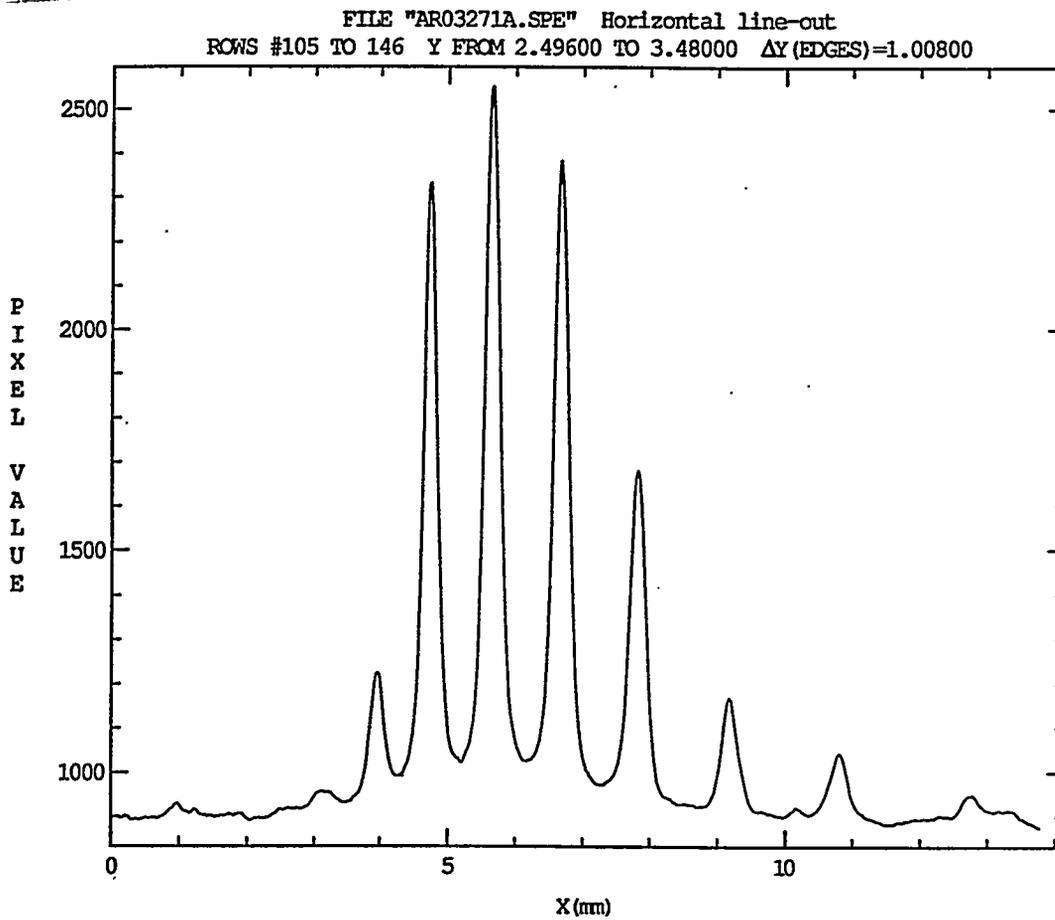
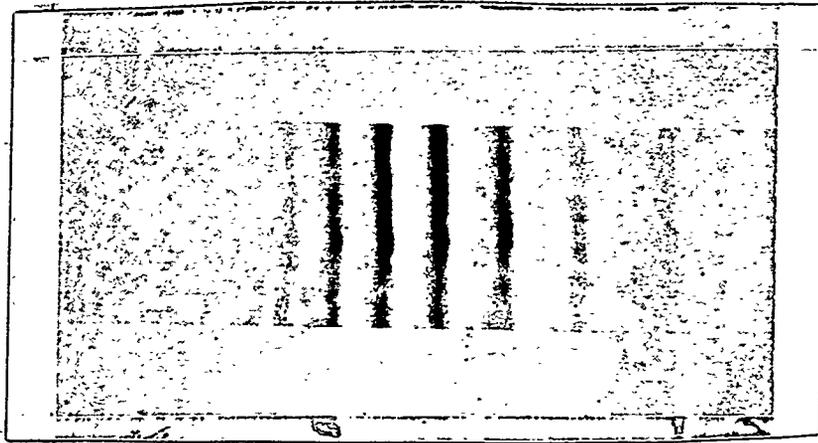


Fig. 2 The x-ray spectrum generated with Ar gas and the planar gas jet.

the harmonic generation yield goes down. In addition, an elliptically polarized laser beam (as opposed to linear polarization) minimizes the harmonic generation. The center of the figure represents approximately the 31st harmonic (26.6 nm) of the 824 nm laser beam.

This experiment was performed during the 1st two days of the allotted experimental period. It was terminated when the planar gas jet poppet mechanism began to malfunction and required engineering modifications.

The next experiment required integrating the streak camera onto the spectrograph in place of the CCD camera. Data was collected in both the integration and time streaked modes using both the planar gas jet and the axially-symmetric gas jets. As with the earlier experiment, both Ar and N<sub>2</sub> gas targets were used. These experiments were performed over the next 3 available days and required lengthy periods of alignment and trouble shooting. A typical time integrated spectrum for an N<sub>2</sub> gas target is illustrated in Fig. 3 where the dominant lines have been identified. The 24.7 nm line represents the transition that Amendt et al<sup>4</sup> suggest might be a candidate for lasing. A typical streaked spectrum for an N<sub>2</sub> gas target generated with the axially-symmetric gas jet is shown in Fig. 4. Similar data was collected for Ar gas targets. The sweep speed on the streak camera was 1 ns/mm. The upper portion of Fig. 4 illustrates a photograph of the streaked spectrum with energy increasing from right to left. Time increases upward in the photograph. The lower portion of Fig. 4 illustrates a time integrated scan along the energy axis. Four slices corresponding to the indicated peak positions were taken along the time axis and are illustrated in Figs. 5 and 6. It appears as though the 20.9 nm line has the fastest recombination rate while the 24.7 nm line has the slowest recombination rate.

The next effort was to perform experiments with the spectrograph/streak camera configuration using a double pulsed laser shot, The laser system was configured

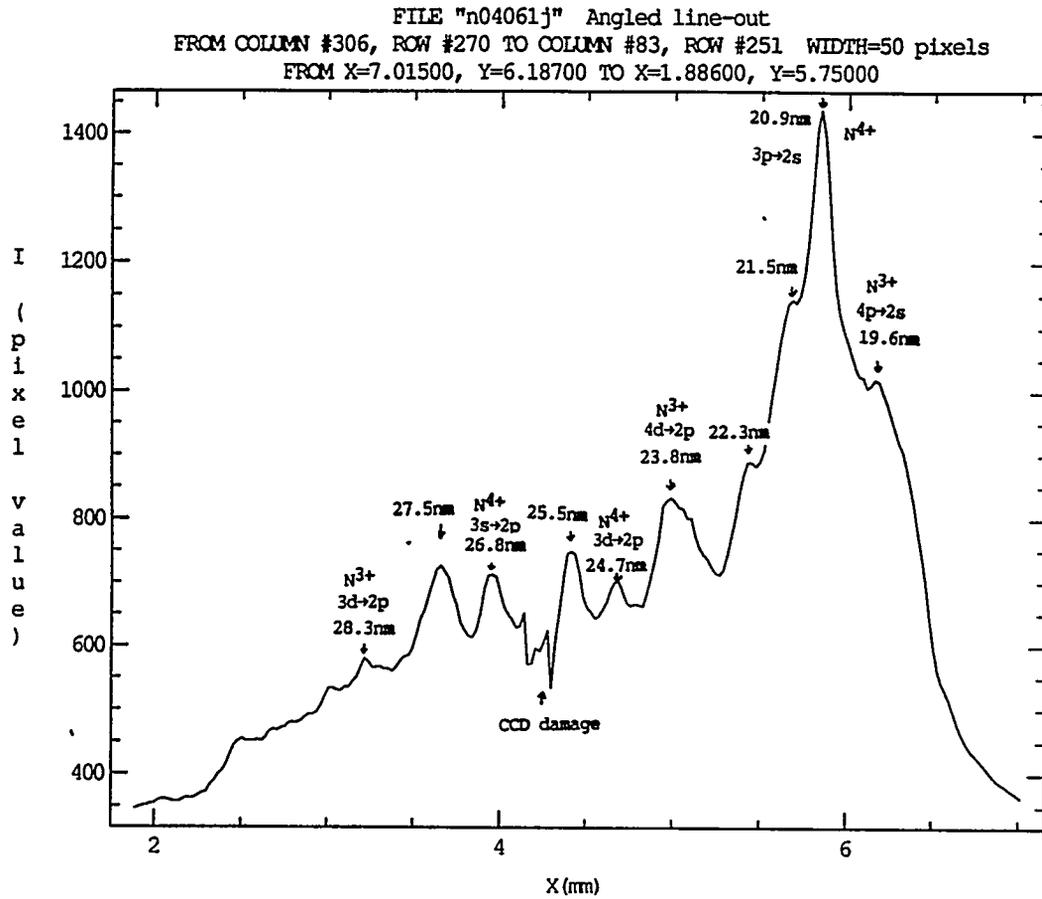


Fig. 3 A typical time integrated spectrum for an N<sub>2</sub> gas target.  
 All of the dominant lines have been identified.

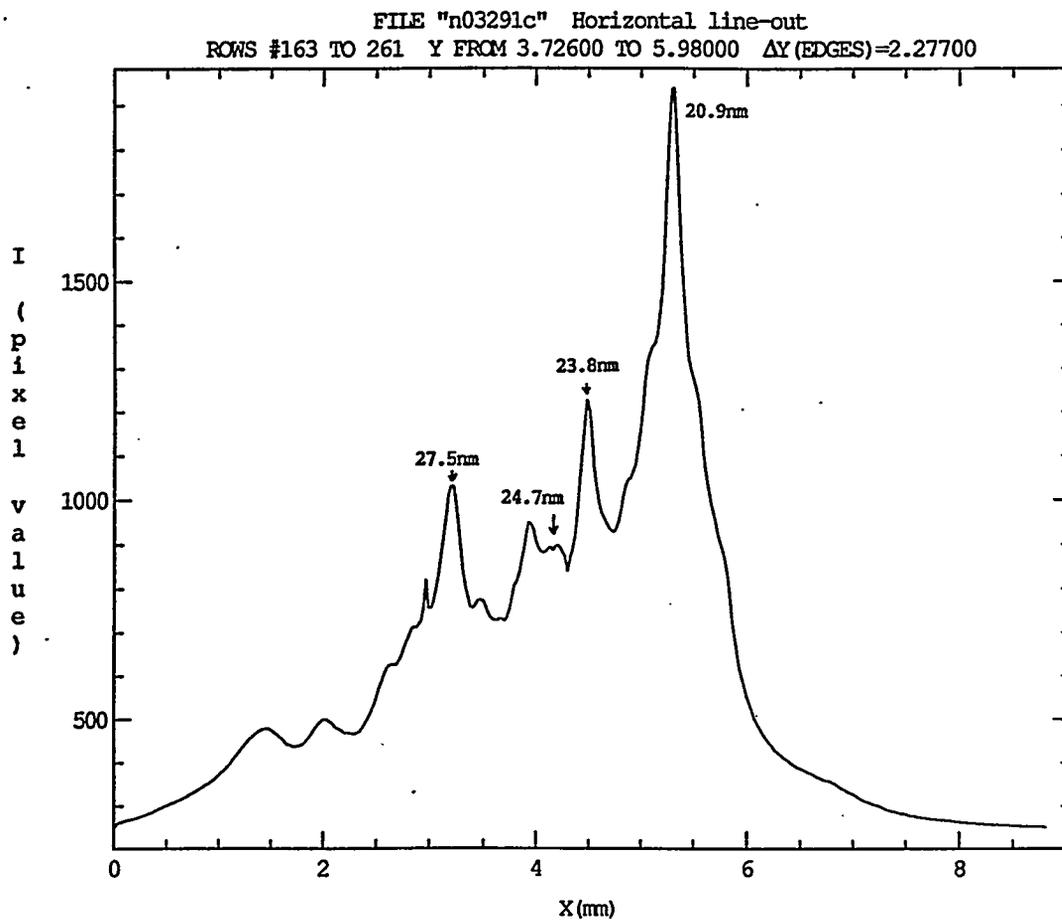
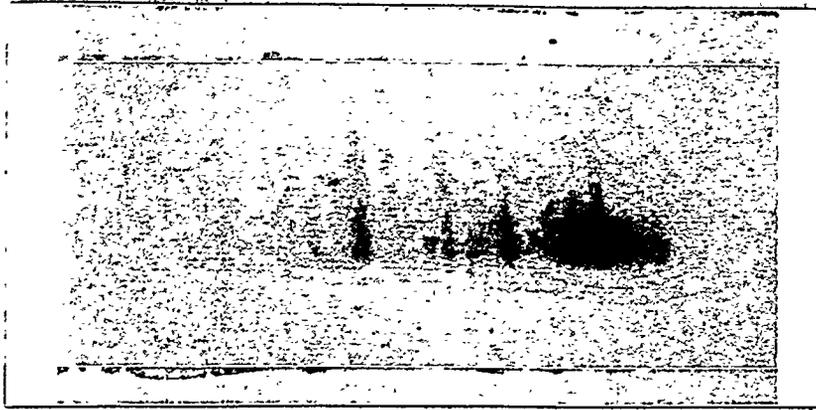


Fig. 4 A typical streaked x-ray spectrum for an  $N_2$  gas target using an axially-symmetric gas jet.

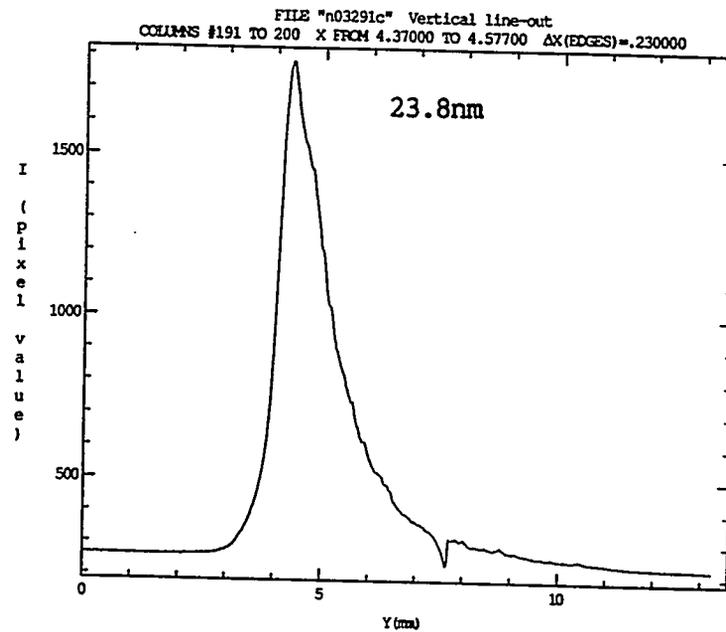
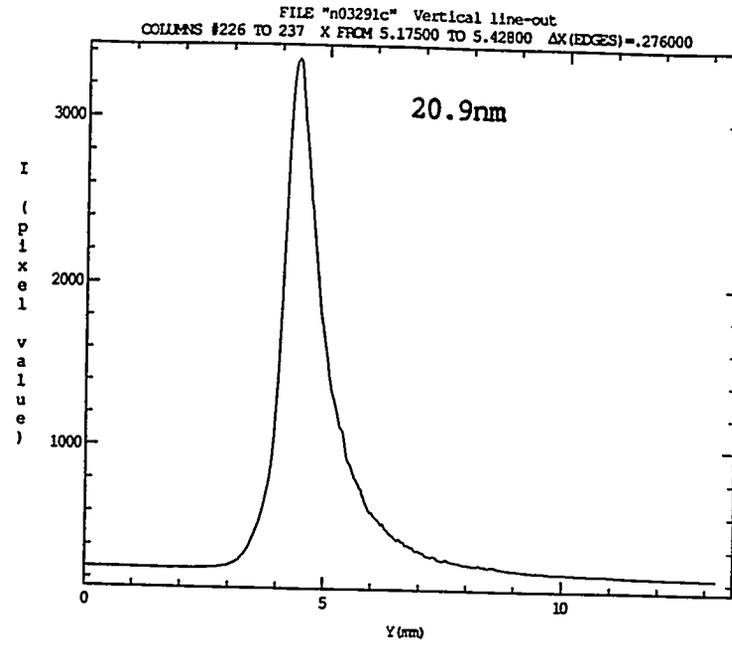


Fig. 5 Scans along the time axis for the indicated energy axis positions.  
The sweep speed is 1 ns/mm.

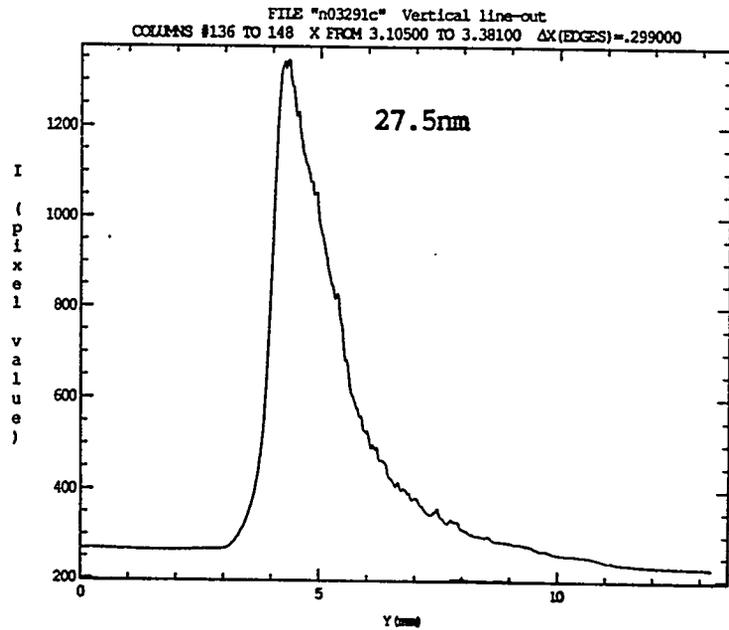
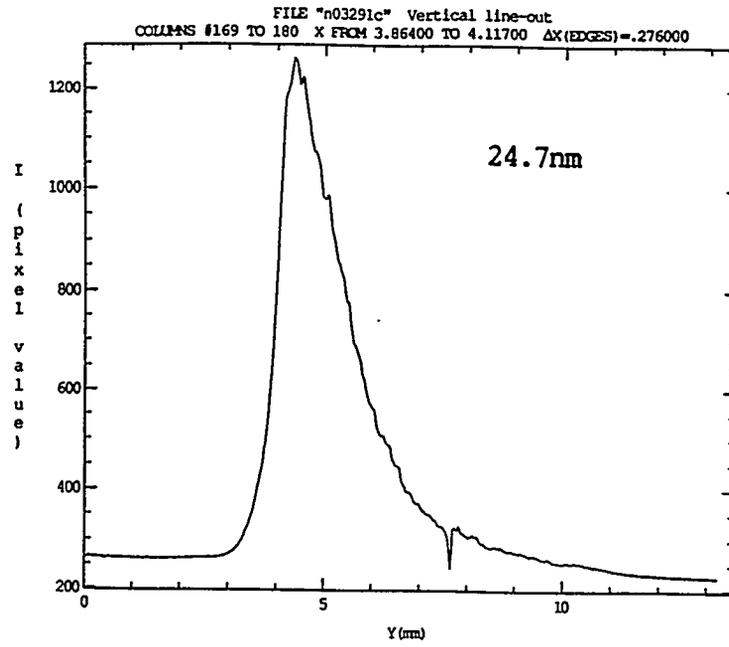


Fig. 6 Scans along the time axis for the indicated energy axis positions.  
 The sweep speed is 1 ns/mm.

to produce a prepulse where 25% of the energy was in the first pulse which preceded the main pulse by 14 ns. Fig. 7 illustrates a photograph of a typical temporal beam profile as measured by the beam monitor. The pulse areas represent the relative energies in each peak and the time scale is 5 ns per large division on the photograph. Experiments were performed for total beam energies between 135 and 210 mJ for N<sub>2</sub> gas targets using the axially-symmetric gas jet and backing pressures of approximately 500 psig. Figs. 8 and 9 illustrate two examples of spectra collected with and without a laser beam prepulse in the non-streaked mode (time integrated). The wave length range is approximately that illustrated in Fig. 3. The upper spectrum in each figure was collected without a prepulse while the lower spectrum in each figure was collected with a prepulse. In each case there is clearly a relatively strong spectrum generated with no prepulse while a very weak spectrum is generated with a prepulse. Note that the intensity scales are different between the upper and lower curves. The sharp peaks between 2 and 4 mm in the upper curves of each figure are due to a damaged region on the CCD image plane. It is thought that the prepulse may be destroying clusters in the jet thereby removing the thermal contribution to the spectrum as compared to when there is no prepulse. In other words, the prepulse prepares the gas so that it acts more like a pure gas jet with no clusters.

Although a significant attempt was made, it was not possible to acquire a video record of the planar gas jet plasma, as had been anticipated, due to mechanical difficulties with the jet during that experiment.

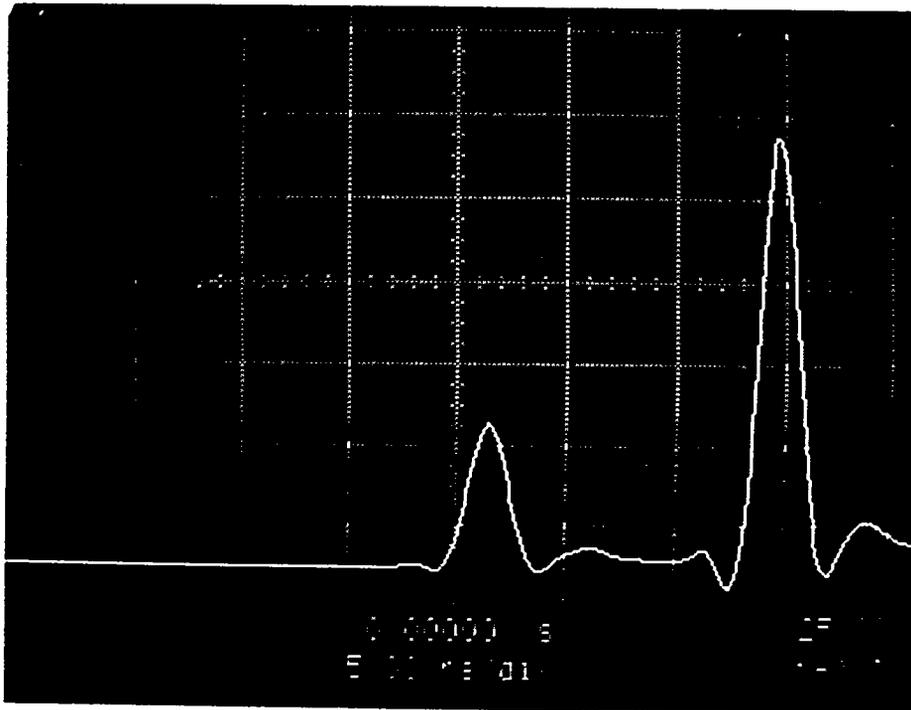


Fig. 7 A photograph of a typical temporal beam profile as measured by the beam monitor during the "prepulse" experiments.

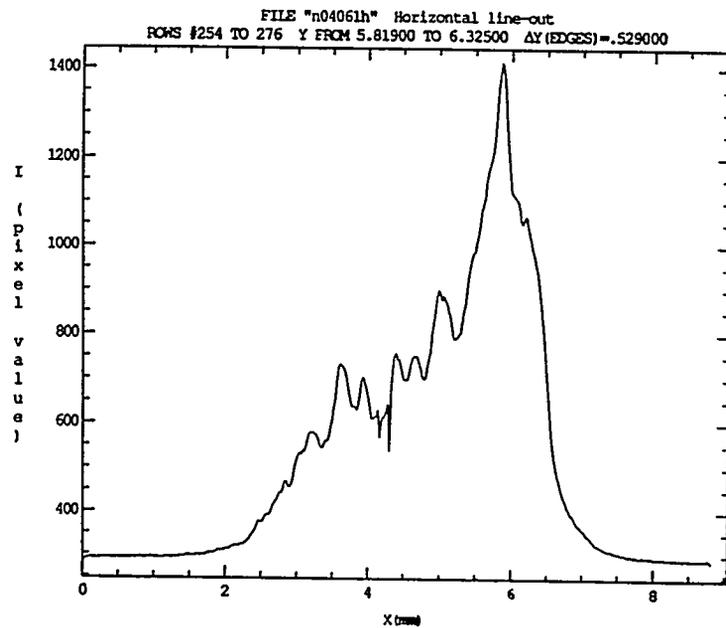
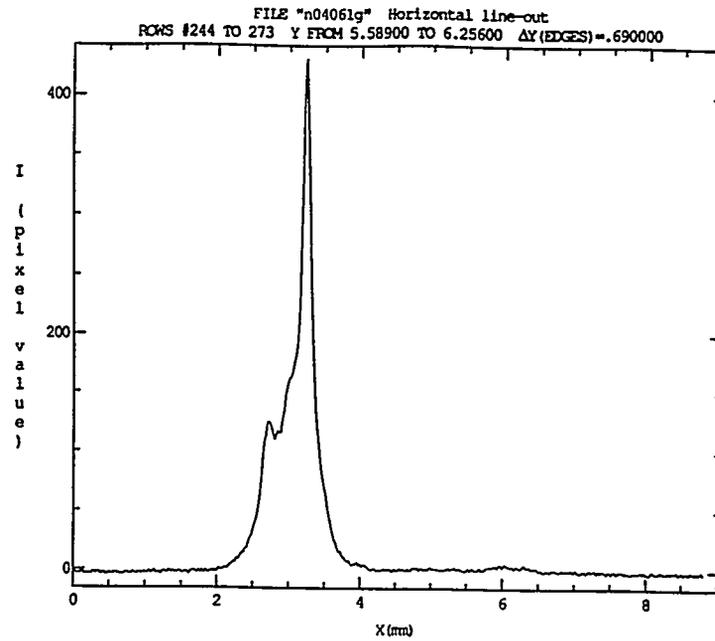


Fig. 8 Energy axis scans of time integrated spectra collected with  $N_2$  gas targets. The upper figure is the spectrum collected with a prepulse while the lower is collected without a prepulse.

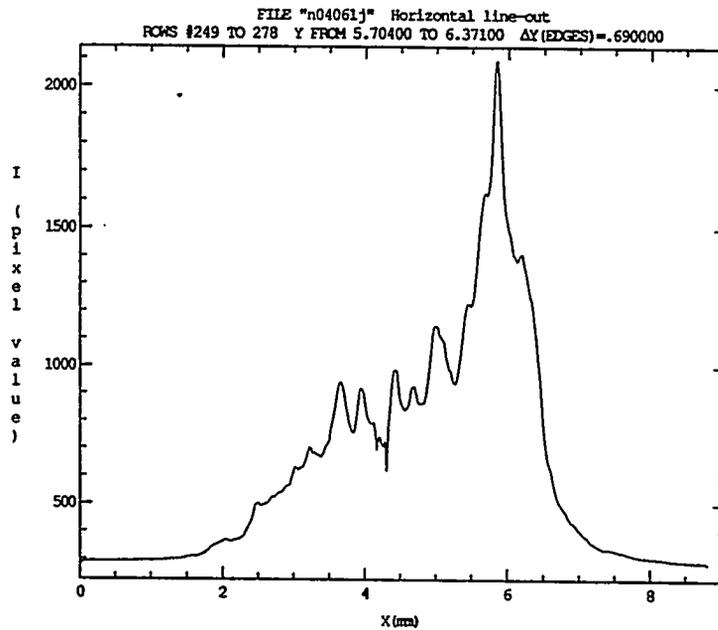
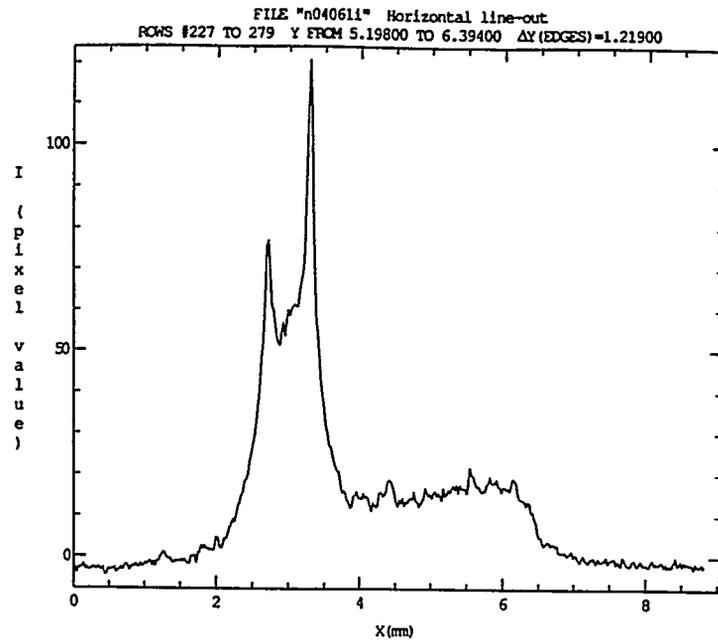


Fig. 9 Energy axis scans of time integrated spectra collected with  $N_2$  gas targets. The upper figure is the spectrum collected with a prepulse while the lower is collected without a prepulse.

## Section 4

## SUMMARY AND CONCLUSIONS

The initial goal of the this experiment was to study and optimize the experimental conditions required for producing lasing for atomic transitions at x-ray energies in  $N_2$  gas target plasmas using ultra-short laser pulses. Amendt et al<sup>4</sup> outlined the conditions for which x-ray lasing might take place for certain defined transitions through the optical-field-ionization (OFI) process. In particular, they outlined the conditions for which x-ray lasing in the 24.7 nm Li-like nitrogen ( $N^{4+}$ ,  $3d \rightarrow 2p$ ) transition might occur. The ultimate goal, after optimization of the excitation and experimental conditions, was to measure the lasing gain using a planar gas jet target.

A series of experimental procedures were initially performed which optimized conditions for the generation and identification of the 24.7 nm x-ray. These included exploring the timing relationship between the laser beam and the creation of the gas jet target, configuring and aligning spectrographs, and optimizing target gas pressures. A number of lines were observed and the transitions identified using the spectrograph in streak camera mode which had been calibrated using harmonic generation and Ar spectral data. Streak camera data suggested that the recombination times might be slightly longer for the 24.7 nm transition compared to other observed transitions. This could imply that the cascading to the  $N^{4+}$  3d state may be more complicated than that to other states.

It is not clear from the experiments described in this report whether the 24.7 nm transition is generated purely through the OFI process or whether some of it is due to thermal heating of gas jet clusters. Experiments performed using the laser in prepulse and non-prepulse mode suggested that spectra generated might

be from heating clusters in the gas jet. This is based on the fact that very weak spectral data is generated with a prepulse as compared to the non-prepulse mode. On the other hand it may be that the plasma degenerates during the 14-ns interval between the two pulses.

The original concept was to use the planar gas jet and line focussed beam pulses to test for lasing gain. Unfortunately, a series of mechanical difficulties were encountered with the poppet mechanism on the planar gas jet during this experiment. Consequently, most of the data collected in this experiment were obtained with axially-symmetric gas jets. It is possible to improve the reliability of the planar gas jet for future experiments, however, it may not be possible to work with cm long plasmas due to refraction and scattering of the beam out of the plasma over those lengths. The path length of the planar gas jet could be controlled via a variable slot through which the gas must pass to reach the region where it intersects the beam. However, it may be that the mm dimensions of the axially-symmetric gas jets may suffice when attempting to measure lasing gain even though the density is not as uniform as in a planar gas jet.

Ultimately the ideal configuration for setting up the required lasing conditions would make use of a planar gas jet with a line focussed driver laser beam entering the gas jet at some shallow angle with respect to the long dimension of the gas jet cross section. In this approach, the ionizing laser beam does not have to traverse the full length of the potential x-ray gain path, thereby reducing the deleterious effects of refraction on the laser beam propagation. The shallow angle would have to be adjusted so that the phase mismatch accumulated between the resultant x-ray and the driver beams over the length of the x-ray gain path would be less than the driver pulse length or the gain peak width, whichever is the greater. For instance, with a gain path length of ~ 1 cm (as in the planar gas jet developed for this study) and a beam angle of the order of a few degrees the phase between the driver and the developing x-ray

beams could be adequately matched. It should be noted that this matching condition was not relevant for the original proposal because the time scales for that mode of x-ray laser generation were much longer and normal entry of the driver beam would be a natural choice leading to the optimum reduction of the effects of refraction in the plasma.

Due to the limitations in the access to the laser facility, there are a series of optimization procedures that were not completed and should be included in any subsequent experimental study. These include the optimization of laser intensity and gas jet pressure as well as laser spot size and beam focal properties.

It is not clear that the present experiment produced conditions in which the proposed lasing scheme could be tested. Additional experiments are required and further optimization, as discussed above, should be carried out before attempting gain measurements.

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