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CONF-880904-10

PREPARED FOR THE U.S. DEPARTMENT OF ENERGY, UNDER CONTRACT DE-AC02-76-CHO-3073

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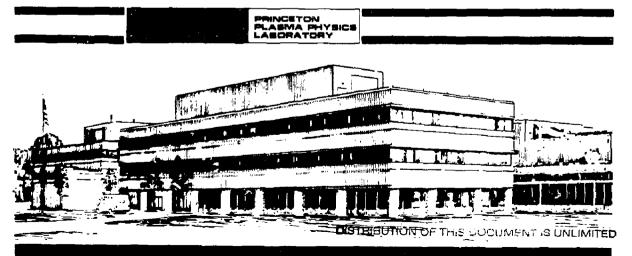
X-RAY LASER RELATED EXPERIMENTS AND THEORY AT PRINCETON

BY

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APRIL 1989

MASTER



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X-Ray Laser Related Experiments and Theory at Princeton*

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ABSTRACT

This paper describes a new system for the development of an X-ray laser in the wavelength region from 5 nm to 1 nm utilizing a Powerful Sub-Picosecond Laser (PP-Laser) of expected peak power up to 0.5 TW in a 300 fs pulse. Soft X-ray spectra generated by the interaction of the PP-Laser beam with different targets are presented and compared to the spectra generated by a much less intense laser beam (20-30 GW). A theoretical model for the interaction of atoms with such a strong laser EM field is also briefly discussed. The development of additional amplifiers for the recombining soft X-ray laser and the design of a cavity are presented from the point of view of applications for X-ray microscopy and microlithography. This overview concludes with the presentation of recent results on the quenching of spontaneous emission radiation and its possible effect on the absolute intensity calibration of soft X-ray spectrometers.

- Invited talk at the conference on Short Wavelength Coherent Radiation, Cape Cod, Mass., Sept. 26-29, 1988.
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I. Introduction

There has been rapid progress recently at a number of laboratories towards the development of X-ray lasers in wavelength regions shorter than 20 nm and of lasers in the VUV region near 100 nm and below. However, in this paper we do not discuss those works, some of which represent very important achievements -- such as reaching very high gain (much above saturation) in Cesium near 90 nm using relatively low pumping power as demonstrated by the Stanford Group [1], generating very high power density radiation (above 10¹⁸ W/cm²) for application to X-ray lasers by the Chicago Group [2], and the proposal to construct a "tabletop" VUV laser at 30-40 nm using Ni-like or Nd-like ions by the MIT Group [3].

The intention of this paper is to give a short overview and provide an introduction to the experimental and theoretical work at Princeton as presented in a series of five papers [4-8]. The topics of these papers include: (i) the development of a powerful sub-picosecond laser system that uses a new approach in X-ray laser development to generate soft X-ray spectra; (ii) a theoretical model for atoms in very strong EM fields; (iii and iv) improvement of the present 18.2 nm laser and its applications to X-ray microscopy; and (v) some difficulties with the calibration of the soft X-ray spectrometers due to the quenching of Einstein A-coefficients.

II. Approach Toward a 1 nm X-ray Laser

The main difficulty in approaching shorter and shorter wavelengths is the requirement for very large increases in pumping power. For example, for X-ray lasers

pumped primarily by recombination or electron-excitation processes, the pumping power P is proportional approximately to λ^{-4} for constant gain g. This follows from a simple relation between gain, wavelength λ , and population inversion ΔN_{inv} (see e.g. [9]).

$$g \sim \lambda^{4} \Delta N_{inv}$$

while

Therefore, in order to decrease the lasing wavelength from 10 nm to 1 nm, the pumping power must be increased approximately by a factor of 10⁴.

The laser presently operating at 18.2 nm requires a laser pumping energy of ~300 J. Without changing pulse length the energy for lasing at 1 nm would have to be on the order of tens of MJ. Because the size and cost of a laser increase dramatically with energy (but not with power), such a system would be very large and very expensive. Thus, a great deal of attention in X-ray laser development is devoted to schemes in which the metastable and autoionizing levels can be used for storing pumped energy, as proposed by S. Harris [10], or schemes based on very short (picosecond and sub-picosecond) pumping pulses [11]. This last approach is

particularly attractive because by decreasing the wavelength of the lasing transition, the lifetime of the ion in the upper state $\tau=1/A$ decreases as λ^2 (A is the spontaneous transition probability), and is of the order of 10^{-12} - 10^{-13} s for a transition wavelength of ≈ 1 nm. Therefore, pumping is required only for a picosecond or sub-picosecond time duration. After this time energy would just be wasted in heating the target material.

Lasers with beam energy of order of only 1 J and pulse duration of 1 ps can provide very high power, $P \sim 10^{12} \, \text{W}$. Even more importantly, such a laser operating in the UV range (e.g., the KrF excimer laser with wavelength 0.25 μ) can be focussed to a 2-3 μ size, providing tremendous power density on the target. The power density can be in excess of 10¹⁸ W/cm² (with corresponding electric field $\sim 10^{11} \, \text{V/cm} = 1 \, \text{kV/Å}$). With this power density it is possible to provide multiphoton excitation and multiphoton ionization of highly ionized ions and use such processes for the creation of population inversion and gain at wavelengths down to 1 nm.

It is very difficult, however, to both create highly ionized ions and provide selective multiphoton excitation of such ions with a single laser. Therefore, our approach uses two lasers [12]. The role of one laser with relatively low power and high energy (e.g., 0.5 kJ, 50 ns CO₂ or 100 J, 3 ns Nd/YLF laser) is to create a plasma column of highly ionized ions which may be confined in a strong magnetic field. The role of the second, extremely high power laser (1 J, 1 ps KrF laser) is to generate gain by multiphoton ionization (MPI), very fast ionization (e.g., inner shell ionization), or selective multiphoton excitation. This approach is illustrated in Fig. 1. More details

about the Powerful Picosecond (Sub-Picosecond) Laser (PP-Laser) system are presented by L. Meixler et al. [4].

In Fig. 2 is shown the experimental arrangement. The same 0.5 kJ CO2 laser (or 100 J Nd/YLF laser), which is used for pumping the 18.2 nm laser [13], is redirected by a system of mirrors and focussed onto a thin foil or fiber target by cylindrical lenses. producing an up to \sim 1-1.5 cm long and 190-150 μ wide line focus. The target is placed in a vacuum chamber inside a 20 cm long solenoidal magnet, designed for magnetic fields of up to 150 kG. Axial radiation from the plasma column in the range 0.5 nm - 30 nm can be measured by the grazing incidence Schwob-Fraenkel spectrometer "SOXMOS" [14]. The core of the experiment is the PP-Laser which is focussed on the plasma column opposite from where the CO2 or Nd/YLF laser is focussed. The PP-Laser can be focussed on the plasma in such a way that travelling wave gain is provided. Synchronization of the lasers, which constitute the PP-Laser system (a simplified block diagram of the PP-Laser is shown in Fig. 3), and time control of the PP-Laser system in relation to the triggering of the CO2 or Nd/YLF lasers. magnet, and diagnostics are provided by several "Fast" and "Master" (slow) electronic timers. (These timers were specially developed in our laboratory for the experiment.) The control system, timers, and data acquisition devices (computers) are located inside a double-shielded Faraday Cage in order to isolate them from strong EM noise coming mainly from the CO2 laser, the large aperture KrF amplifier, and magnet. Most of the signals to and from the Faraday Cage are transmitted by optical fibers.

In Fig. 4 is shown a picture of the PP-Laser system without the final KrF amplifier. Located on the right-hand side of the picture are the mode-locked YAG laser, dye laser, and three-stage dye amplifier pumped by a 3 J Quantel Nd/Glass laser. On the left-hand side are shown two Lamda-Physics KrF amplifiers. A large aperture (5 x 10 cm) KrF amplifier (3rd KrF amplifier) was added to the system and is shown in Fig. 5. This amplifier was developed at Princeton University in cooperation with J. Goldhar (Univ. of Maryland) during the last ~2.5 years and is now operated routinely as part of the PP-Laser system. It is a 100 cm long, fast discharge amplifier, which is operated in a double pass mode. The energy of the seed pulse from the second KrF amplifier is 10-15 mJ and the pulse duration (without compression) is ~ 1 ps. Using a fiber-gratings system for pulse compression (work done in cooperation with J. Fujimoto, MIT), we obtain a pulse duration of 250-300 fs and final amplifier output energy of ~ 150 mJ. In Fig. 6 is shown measurements of the pulse duration using a two-photon excitation technique in Xe [15]. Reference 4 presents more details about the large aperture KrF amplifier as well as measurements of the pulse duration for noncompressed and compressed pulses. Measurements of the beam pulse duration after passing through the final amplifier are not yet available; however, indirect measurements indicate that the final amplifier does not increase the pulse duration, implying a PP-laser beam power of P ~ 0.5 TW. The focal spot of such a beam using a less than optimal f/10 lens is expected to be $\sim 7-8~\mu$ in diameter providing a power density close to 1018 W/cm2 (with a new t/2 or t/3 focussing system the power density should be several times higher).

One of the first experiments to use such a high power density laser beam measured soft X-ray spectra for carbon and fluorine. The laser beam was focussed on a rotating cylindrical teflor target while a soft X-ray grazing incidence Schwob-Fraenkel spectrometer "SOXMOS" with multichannel detector monitored the plasma radiation from the target surface. In the lower part of Fig. 7 is shown the spectrum in the vicinity of the CVI 33.74 Å and CV 40.27 Å lines (both from $2 \rightarrow 1$ transitions). Besides the enormous line broadening, one may see a strongly pronounced, unusual structure in the lines. Both broadening and structure are larger than in the spectra obtained with the 20-30 GW, 1 ps PP-Laser beam (without final KrF amplifier) shown in the upper part of Fig. 7. It should also be noticed that the number of shots needed for these short wavelength spectra is not proportional to the laser beam energy but rather to its power. (The energy of the laser beam increased by a factor of 5-7 while the number of shots decreased by a factor of 20.) In spectra obtained earlier with the 20-30 GW PP-Laser (10¹⁶ W/cm²), part of the large broadening and asymmetry of the FVII lines in the 120-140 Å spectral region was attributed to the Stark effect and radiation from the forbidden components of the lines [16]. Very recently K. Koshelov [17] interpreted asymmetric broadening of the FVII lines to be a result of satellite line radiation. Spectral lines of CVI, CV, and FVII (Figs. 7,8) excited by the very high power beam seem to indicate a complicated satellite-type structure. Of course, the very strong electric field created by such a laser beam may be responsible for these effects. Spectroscopic data for different targets as well as experiments in which a highly ionized plasma is initially generated by CO2 or Nd/YLF lasers and then excited by the

PP-Laser should enable us to develop a clearer picture of the behavior of highly ionized ions in strong laser fields.

In the meantime, theoretical work continues on the basic questions related to the use of the PP-Laser for creating population inversions in the region 5-4 nm to 1 nm. One such question is: how is the ratio between the rate of excitation of the chosen level n and the rate of ionization from this level dependent on the laser intensity (multiphoton processes) and Z of ions? Another question for consideration is: does the presence of other bound states influence the ionization from a given bound state?

The theoretical model is based on a sequence of δ -function-confining potentials as an approximation to a continuous potential. Such a model allows for relatively simple analytical and numerical calculations of the multiphoton ionization rate under strong field conditions. Some of the results are presented by S. Susskind et al. [8].

III. Progress on the Recombining Soft X-Ray Laser

(a) Development of an Additional Amplifier at 18.2 nm

Presently, the highest beam energy of our X-ray laser [18] at 18.2 nm (CVI 3-2 transition) pumped by a $W_L \approx 300$ J CO₂ laser in a 90 kG solenoidal magnetic field is $W_X \approx 3$ mJ with a 3 min repetition rate. A higher repetition rate is predicted for a recently designed commercial prototype soft X-ray laser (SXL) with a superconducting coil. This system would also be more compact than the SXL at PPPL. At present the beam energy W_X is one of the most important parameters for applications of the SXL.

In order to increase W_X we have developed an additional SXL amplifier (3 mm long) at 18.2 nm that is pumped by a line-focussed Nd/YLF laser beam onto a carbon target. In Fig. 9 the arrangement of the CO_2 -laser-pumped SXL with two additional SXL amplifiers inside the solenoidal magnet is shown schematically. (The magnetic field is crucial for the CO_2 -laser-pumped SXL but it is much less important for SXL amplifiers pumped by the Nd/YLF laser.) Located on the left-hand side of the system is the CO_2 laser beam which is focussed on the center of a four-carbon-blade carbon disc target. Along the path of the soft X-ray laser beam are located two cylindrical carbon targets whose length can be changed from 1 mm to 3 mm by rotation. Thin iron blades in the front of the targets provide additional radiation cooling of the plasma column. We have demonstrated gain up to $g \approx 8 \text{ cm}^{-1}$ in one SXL amplifier with 25 J of Nd/YLF laser beam energy (see D. Kim et al. [6]).

In Fig. 10 are shown axial spectra in the vicinity of the lasing line CVI 18.2 nm (3-2 transition) and in the vicinity of CVI 13.5 nm (4-2 transition) for 1, 2, and 3 mm long targets. One may see a strong nonlinear increase in intensity of the 18.2 nm line (Fig. 11) in comparison to near-linear increase in intensity of the 13.5 nm line. (Note that this line should be optically thin due to the expected population inversion between levels 4 and 2. Also, in the axial spectrometer there is no contribution of the 4th order of the CVI 3.37 nm line to the intensity of the 13.5 nm line.) A significant problem here is to match the transverse size ("aperture") of the SXL amplifier to the original SXL. We were able to create significant gain with only 6 J of pumping energy from the Nd/YLF laser; however, in this case the "aperture" of the SXL amplifier was too small.

We have not yet been able to match the SXL laser temporally or spatially with the SXL amplifier. This may be due to jittering of the CO₂ laser and difficulty in the alignment of the SXL amplifier along the SXL beam path in vacuum.

(b) <u>Cavity</u>

A laser cavity can increase the brightness of the SXL beam by several orders of magnitude by decreasing the divergence to close to the diffraction limit. In order to establish the proper cavity modes, a number of passes through the gain medium are needed and a relatively long duration gain is necessary. The Princeton SXL at 18.2 nm with a gain duration of 10-30 ns seems to be well suited to cavity development.

In our early work, using a newly developed multilayer mirror [19], we demonstrated for the first time a 120% increase in 18.2 nm radiation due to amplification of stimulated emission by using a mirror with reflectivity of only 12% [14]. However, the mirror alignment posed tremendous difficulties and made it practically impossible to use a cavity in the original SXL setup. We, therefore, designed an unstable resonator-type cavity [20] with a transversely pumped carbon fiber as the lasing medium (Fig. 12). A specially designed fiber transport mechanism will enable the use of 15 fibers without opening the vacuum chamber. For the spherical multilayer cavity mirrors a small vacuum chamber was built in which the position of the mirror can be remotely adjusted with high precision. The same cavity will also be used for a 1 cm long SXL created by a line-focussed Nd/YLF laser beam incident on the cylindrical carbon target described in section (a).

In the cavity design, particularly in choosing distances between lasing medium and mirrors, we were concerned with the possibilities of damaging the multilayer mirrors by X-ray radiation. Recently, however, Ceglio et al. [21] demonstrated, in a very elegant cavity experiment, that such mirrors are quite stable against soft X-ray beam damage, even at a distance of a few centimeters from the lasing medium.

(c) Gain in Li-Like lons at 15.4 nm and 12.9 nm

Pioneering work for Li-like AlXI ions, particularly for the 5f-3d transition at 10.5 nm, was done by P. Jaegle and his group (see e.g. [22]) using a Nd/Glass laser (initial plasma electron density $N_e = 10^{21}$ cm⁻³ for the pumping lasing medium. In our system with a CO_2 pump laser the critical electron density is $N_e = 10^{19}$ cm⁻³. For such an electron density, the largest gain in AlXI and SiXII is expected for the 4f-3d transition at 15.4 nm and 12.9 nm, respectively.

The aluminum or silicon targets used in the experiment were very similar to the SXL carbon target with the exception that the blades were a combination of lasing element (Al or Si) and fast radiator (Fe). The measured one-pass gain was $gl \approx 3-4$ for 15.4 nm and $gl \approx 1-2$ for 12.9 nm radiation. Details about the experiment and theoretical modeling are presented in [5].

It is worthwhile noticing here that Li-like ions provide a more efficient lasing medium than H-like ions. In Fig. 13 is shown the lasing wavelength of H-like (3-2 transition) and Li-like (4f-3d transition) ions versus the ionization energy of these ions. Because the required pumping energy increases with ionization energy, lasing action

in Li-like ions can be generated at a shorter wavelength than in H-like ions with the same pumping energy, particularly if one considers 5t-3d transitions.

IV. Applications of the Soft X-Ray Laser

The 18.2 nm laser (SXL) has been used for X-ray contact microscopy of biological specimens. This work is also closely related to X-ray microlithography, about which we are very much interested. The ultimate goal of our X-ray laser microscopy program is to obtain images of living cells, preferably holographical images with three-dimensional reconstruction. As a first step in this direction, we have built a simple contact microscope with help from J. Kirz (Univ. of Stony Brook) and D. Sayre (IBM). The details of this work as well as our other X-ray laser recroscopy works are described in the paper by D. DiCicco et al. [6].

In the X-ray laser microscope, a thin (~0.1 μ) silicon nitride window 0.2 mm x 0.2 mm square separates the vacuum tube, in which X-rays travel, from the biological cells located on photoresist at atmospheric pressure (see e.g. [6, 23]). An aluminum filter less than 0.1 μ thick, with the highest transmissivity near 18.2 nm, prevents visible and UV radiation from interacting with the photoresist. The X-ray laser beam was directed towards the microscope and focussed to a 1 mm x 0.8 mm spot by an ellipsoidal grazing incidence mirror. We demonstrated in this experiment that the SXL beam has sufficient energy to expose photoresist in a single shot. After the initial experiments in which an image of 100 mesh was obtained on PMMA photoresist, images of diatom fragments (the silicified skeleton of planktonic algae) were also recorded on

photoresists. In Fig. 14 is presented such an image on photoresist viewed with a scanning electron microscope (SEM). The picture indicates the resolution on the photoresist is better than 0.1 μ . One may also regard diatom fragments as a kind of lithographic mask and Fig. 14 as an illustration of the potential application of the SXL to microlithography.

In the next step, we built a Composite Optical/X-Ray Laser Microscope (COXRALM) shown schematically in Fig. 15 and described in [6]. COXRALM allows a biologist to select and observe biological cells using an optical phase contrast microscope [24]. The cells are located directly on an optically transparent photoresist. After selection and initial observations, the images of the cell are created on photoresist with the SXL beam in order to obtain higher resolution than with an optical microscope. In this procedure the X-ray laser tube with the 0.1 μ silicon nitride window on its tip is lowered until contact with the cell is made. The photoresist is exposed by triggering the SXL. In Fig. 16 is shown a photograph of COXRALM oriented horizontally (COXRALM can also be in the vertical position) on the SXL system (the lasing medium of SXL is ~300 cm away, on the left).

In Fig. 17, as an example, is shown an SEM image of the replica of dehydrated hela cells (Helen Lane cervical cancer cell) obtained from the Biology Dept. of Princeton University.

Presently our work is concentrated on the preparation of experiments with live cells in a wet environment and on the design of an Imaging X-Ray Laser Microscope (IXRALM).

V. <u>Effect of Quenching Spontaneous Emission Radiation on the XUV</u> Spectrometer Calibration Using the Branching Ratio Technique

The intensity ratio of two spectral lines from the same upper level in an optically thin plasma is equal to the ratio of the respective spontaneous emission, A, coefficients (Einstein coefficients). If the wavelength of one line is in the visible spectral region and the wavelength of the second line is in the VUV or XUV region, by measuring the ratio of the signals of these lines and knowing the absolute intensity of the first line, the absolute intensity of the XUV line can be obtained from the ratio of the A-coefficients. The technique is known as the branching ratio technique for the calibration of VUV and XUV spectrometers [25] and is based on the assumption that the A-coefficients are independent of plasma conditions. Using this technique for the calibration of XUV spectrometers for the X-ray laser experiment, we discovered that the ratio for some A-coefficients, in sufficiently dense plasmas, differed significantly from the ratio in lower density plasmas. Many series of very precise measurements of the ratio of A-coefficients for CIV 5801-12 Å (3p-3s)/312 Å (3p-2s) lines confirmed that first observation [26]. Recently, similar measurements of NV 4603-20 Å (3p-3s)/209 Å (3p-2s) and CIII 5696 Å (3d-3p)/574 Å (3d-2p) lines showed the same phenomena. In Fig. 18 are shown the intensities and intensity ratios for CIII. More extensive and detailed information about these measurements are presented by Y. Chung et al. [17]. In [17] there is also a discussion of theoretical approaches to the problem. However,

up to now we do not have a model which provides a convincing explanation for the experimentally observed quenching of spontaneous radiation.

Acknowledgments

The presented data were the results of very dedicated work by members of the scientific and technical staff and graduate students of the X-Ray Laser Project whose names, as authors, appear in References [4-8]; by visiting scientists, particularly P. Lemaire, J.L. Schwob, and G. Umesh; and a number of scientists associated with us, among them, C. Clark, P.C. Cheng, J. Fujimoto, E. Kohen, R. Kulsrud, M. Littman, T. McIlrath, R. Miles, and C. Oberman. I would like to thank H. Furth for helpful discussions and stimulation of the work on applications of the SXL.

The work was possible due to financial support by the U.S. DOE Advanced Energy Projects of Basic Energy Sciences Grant No. KC-05-01 and the U.S. Air Force Office of Scientific Research Contract No. AFOSR-86-0066.

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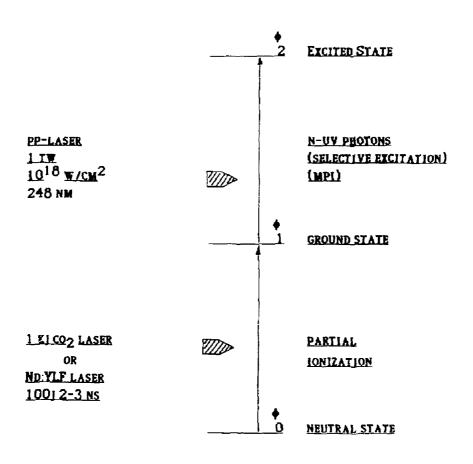
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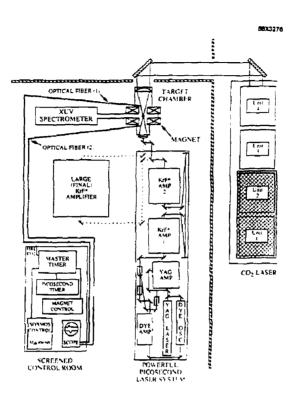
Figure Captions

- Fig. 1: Basic idea of "Two-Laser Approach" to X-ray lasers: a relatively low power and high energy laser "is preparing" ions and an extremely high power sub-picosecond laser creates a population inversion.
- Fig. 2: Scheme of the experimental arrangement for the "Two Laser Approach."
- Fig. 3: Simplified block diagram of the PP-Laser System.
- Fig. 4: Photo of the PP-Laser System without the final KrF amplifier.
- Fig. 5: Photo of the large aperture KrF amplifier (final amplifier KrF III, in center of laboratory), magnet (far left), and vacuum system.
- Fig. 6: Autocorrelation trace of a compressed pulse after the second KrF* amplifier.
- Fig. 7: Spectra obtained (a) without compression and (b) with compressed pulse and final KrF* amplifier.
- Fig. 8: Spectra of FVII obtained with compressed pulse and with final KrF* amplifier.
- Fig. 9: Scheme of arrangement of CO₂-laser-pumped X-ray laser (SXL) with two additional SXL amplifiers.
- Fig. 10: Axial spectra in vicinity of CVI 18.2 nm lasing line and CVI 13.5 nm line for 1, 2, and 3 mm long targets.
- Fig. 11: Gain measurement for CVI.
- Fig. 12: Unstable resonator-type cavity for the SXL.
- Fig. 13: Lasing wavelengths of H-like and Li-like ions versus the ionization energy.

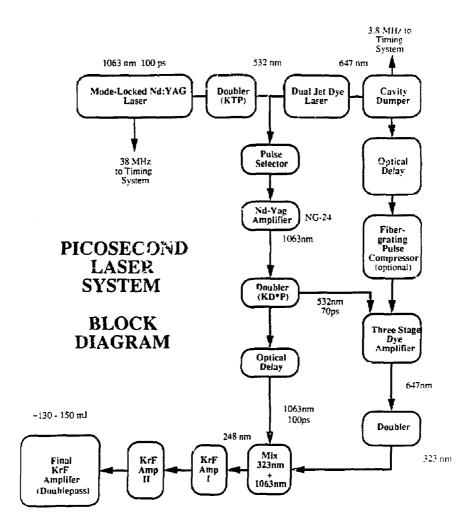
- Fig. 14: Image of diatom fragments (silicified skeleton of planktonic algae on PMMA photoresist viewed with scanning electron microscope (SEM).
- Fig. 15: Scheme of Composite Optical X-Ray Laser Microscope (COXRALM).
- Fig. 16: Photo of horizontally oriented COXRALM attached to the SXL.
- Fig. 17: SEM image of a replica of hela cells (Helen Lane cervical cancer cells).
- Fig. 18: Relative intensities and branching ratio for two CIII lines.

TWO LASER APPROACH TO LASING IN 10 Å REGION





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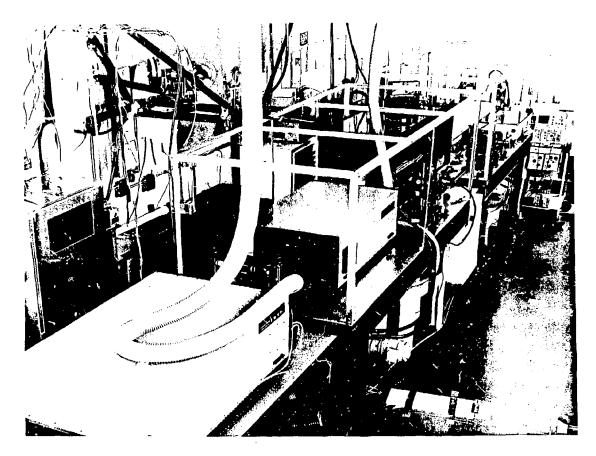


Fig. 4

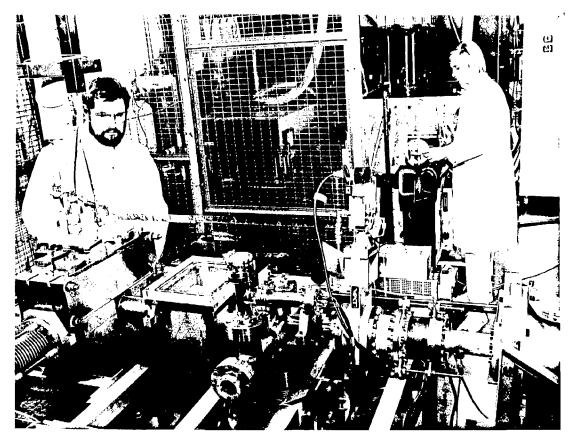


Fig. 5

88X3273

COMPRESSED PULSES

Output from KrF Amp II of PP-laser (Two photon fluorescence technique in Xe)

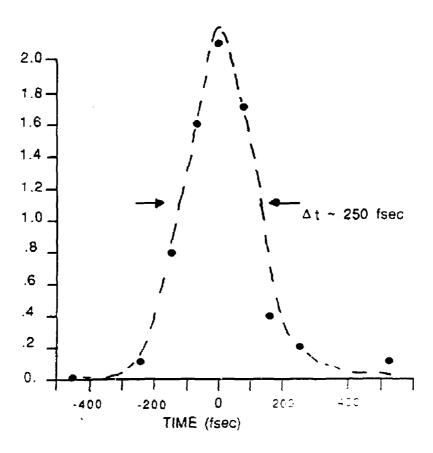


Fig. 6



35

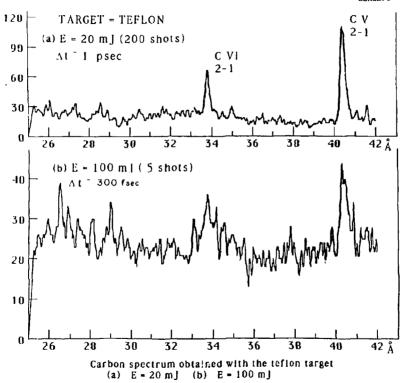


Fig. 7

Fig. 8

Two Amplifier Experiment

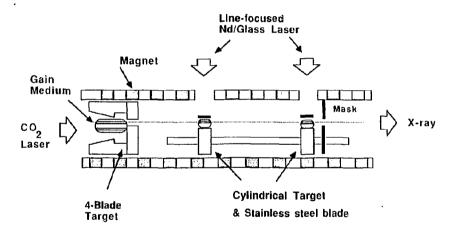
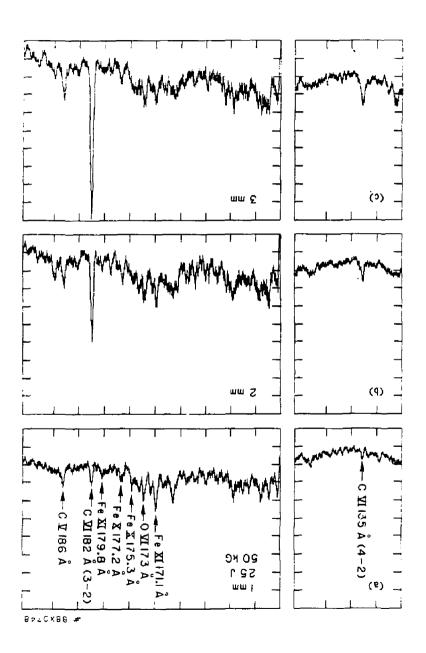


Fig. 9

t



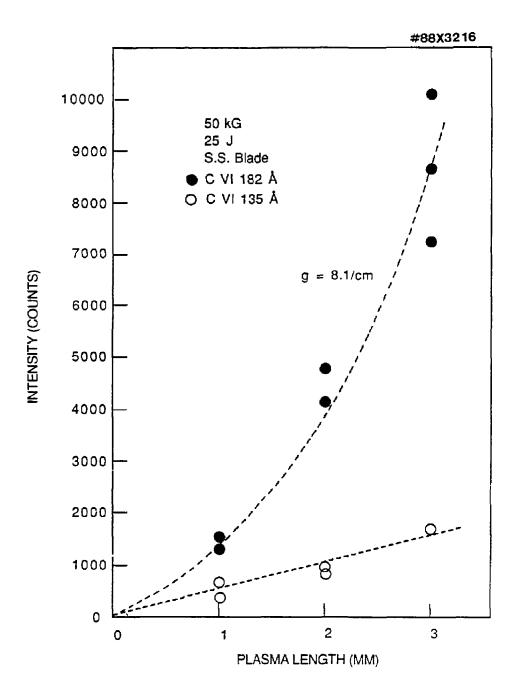
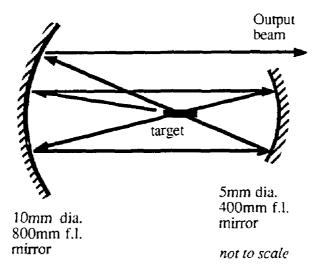


Fig. 11

88X3274



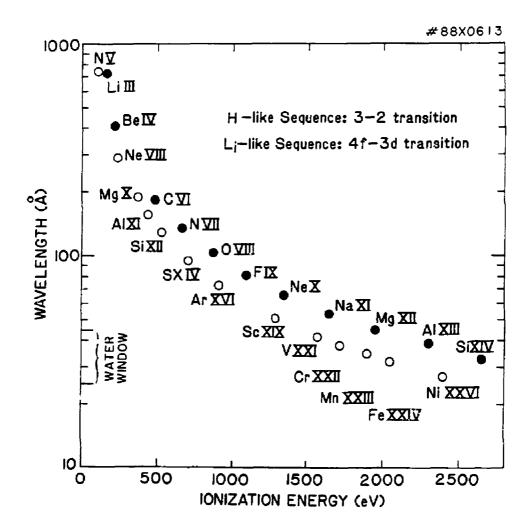
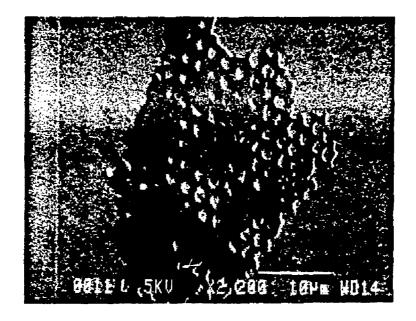


Fig. 13



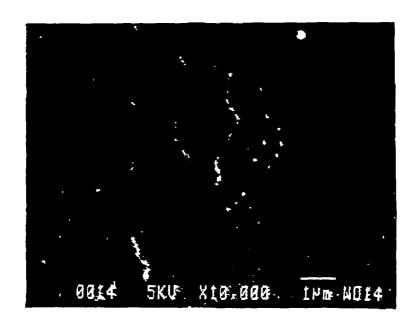


Fig. 14

87X0675

COMPOSITE X-RAY LASER MICROSCOPE (COXRALM)

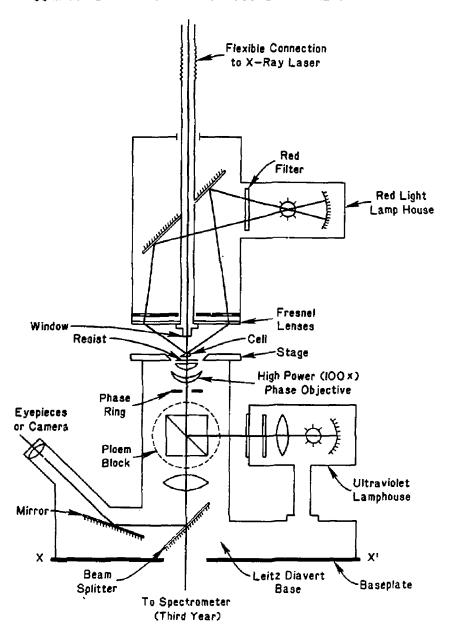


Fig. 15

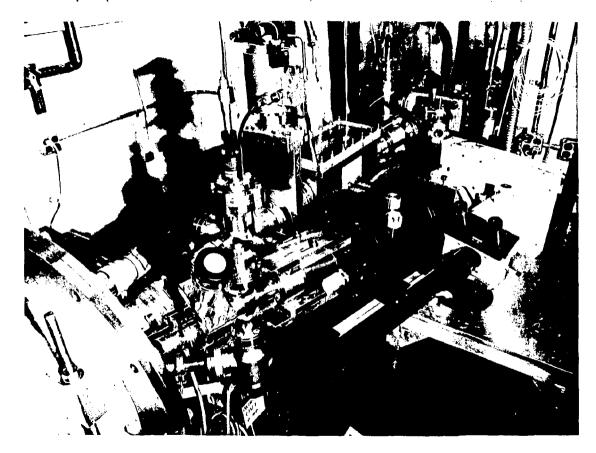
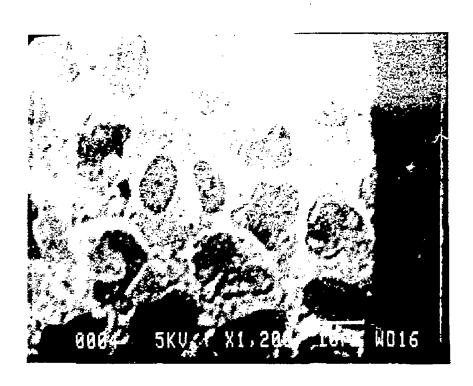
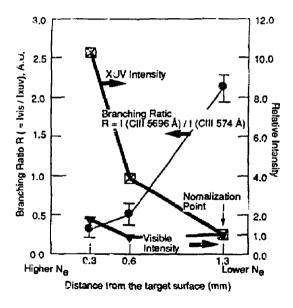


Fig. 16





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