Toward Shorter Wavelength Lasers and Soft X-Ray Laser Microscopy


Abstract—We present two approaches to X-ray laser development at Princeton and review progress toward the wavelength region below 10 nm. In addition, we present the first results from the application of the existing soft X-ray laser at 18.2 nm to X-ray microscopy.

I. INTRODUCTION

FOLLOWING the first joint announcement by groups at Princeton University and Lawrence Livermore National Laboratory at the American Physical Society Meeting in Boston in November 1984, progress in increasing the output power and range of soft X-ray lasing wavelengths has been rapid [1]-[7]. These sources are beginning to be used in several applications. In this article we will review the progress toward soft X-ray laser development in the wavelength region below 10 nm at Princeton and present the first results from the application of a soft X-ray laser to microscopy.

II. THE PRINCETON SOFT X-RAY LASER

We first review the Princeton soft X-ray laser at 18.2 nm, in which a population inversion and gain are generated in a rapidly recombining plasma confined in a magnetic field. Gain at shorter wavelengths has also been measured using lithium-like ions, i.e., AlXI (15.4 nm) and SiXII (12.9 nm) (discussed below). A commercial CO2 laser (maximum energy 1 kJ, duration 70 ns) is focused onto a carbon disk located in a strong (up to 90 kG) magnetic field, creating a carbon plasma of sufficient temperature that the electrons are stripped off most of the carbon ions. After the laser pulse the plasma cools rapidly by radiation losses and fast three-body recombination preferentially populates the upper excited levels in hydrogen-like carbon, CVI. The m = 2 level is rapidly depopulated by the strong 2 → 1 radiative transition, and in this way a population inversion is built up between level n = 3 and m = 2 leading to amplified spontaneous emission and lasing at 18.2 nm.

The strong confining magnetic field provides several advantages. It enables the electron density to be controlled to the optimum value and forms the plasma to a long thin geometry suitable for a laser. The geometry is also suited to fast radiation cooling, and the radiation cooling can be enhanced by the addition of high-Z materials.

The CO2 laser is focused into a target chamber inside a solenoidal magnet that is surrounded by diagnostic spectrometers (see Fig. 1). The spectrometers are absolutely calibrated and measure the soft X-ray gain by measuring the directionality of the emitted radiation. In a long thin plasma most of the stimulated emission is along the plasma axis and the gain is measured by comparing the axial to transverse intensity.

The target is a carbon disk with one to four blades attached perpendicularly to the target surface. The blades help to generate an elongated plasma. They also help to cool the plasma by thermal conduction and additional radiation losses. An important feature is the radial profile of the plasma. The plasma pressure is balanced by the magnetic field pressure so that on axis there is a high temperature due to heating by the laser and corresponding low density. In the outer cooler regions the density is higher, and it is in these outer regions that the conditions are most favorable for fast recombination. The gain is generated in an annular region around the center of the cylindrical plasma. An off-axis slot in the target transmits the stimulated emission to the axial spectrometer. The system has a very accessible output beam, can be fired every few minutes, and can be operated by only two people.

With carbon targets gain is generated on the 3→2 transition in CVI at 18.2 nm. The 4→2 transition at 13.5 nm emits mostly spontaneous emission, so one way to measure gain is by the axial enhancement of the 18.2 nm emission compared to the spontaneous, isotropic 13.5 nm emission. A second, independent way is to use the spectrometer calibrations to measure the relative intensity in the two directions. Reference [2] describes gain measurements of gL = 6.5 corresponding to an axial enhancement of the 18.2 nm emission of a factor of 100. This is illustrated in the spectra in Fig. 2, showing the very high intensity of the 18.2 nm emission in the axial direction. Measurements of higher values of gain, gL = 8, with enhancement of up to 500 of the 18.2 nm line are presented in [3].

To get an absolute measure of the divergence the axial spectrometer was scanned across the soft X-ray beam. A...
beam divergence of 5 mrad was measured at magnetic fields of 35 and 50 kG [3]. In summary, earlier work at Princeton has resulted in a soft X-ray laser with the following characteristics: 18.2 nm wavelength; 1–3 mJ pulse energy; 10–30 ns pulse duration, and 5 mrad beam divergence.

III. SOFT X-RAY LASER MICROSCOPY

Much progress has been made in biology and medicine due to the high-resolution images obtained from electron microscopes. However, in order to be viewed by an electron microscope the specimen must be dried, stained, and sectioned, and it is clear that some information about the living cell is lost in the process. One can view a live cell with a light microscope with high fidelity but the resolution is, of course, limited. Soft X-ray contact microscopy offers a new method to obtain high-resolution images of live cells. The specimen is contained in an environmental cell isolated from the X-ray laser vacuum system by a 120-nm-thick silicon nitride window. The image is recorded on photoresist which is later viewed by an electron microscope. This approach has been used recently with plasma light sources and synchrotrons [8], [9]. The soft X-ray laser has the advantage of a 10–30 ns exposure time enabling flash images of live cells to be recorded, unlike the several minutes needed for synchrotron sources which effectively prevents the imaging of live cells with synchrotrons. The highly collimated output beam of the soft X-ray laser, compared to a conventional plasma light source, has the advantage of less penumbral blurring of the image and more flexible microscope design.

During microscopy experiments the soft X-ray beam is diverted 20 degrees via an astigmatic spectacle lens which serves as a rudimentary toroidal grazing incidence mirror. Fig. 3 gives a view of the mirror, the positioning system, and the rear portion of the environmental cell. The translators are remotely controlled and allow us to steer the X-ray beam to the environmental cell. The alignment is optimized by temporarily placing a PIN diode detector at the environmental cell position.

Our environmental cell design follows the arrangement used by Feder et al. [9]. A silicon nitride window serves as the vacuum interface. The window is 200 μm × 200 μm × 120 nm thick and is coated with 100 nm of aluminum. The Al acts as an UV rejection filter and also lends some mechanical support to the membrane. Initial experiments have been performed in order to evaluate the performance of the system without the complications involved in handling live specimens [10], [11]. The first of this series used a piece of #100 wire mesh in place of a living cell. Images of this mesh were recorded on Kodak 101 film and on P(MMA co MAA) resist and may be seen in Fig. 4. Both of these images were generated with one laser shot. They differ only in the fact that the resist image was obtained without the use of the aluminum-coated silicon nitride window, so a contribution from UV light from the plasma cannot be ruled out at this time. The P(MMA co MAA) resist was developed in equal parts of methyl isobutyl ketone and isopropanol. Images obtained on resist with the window in place were too faint to be clearly identifiable using a metallurgical microscope, and an effort is being made to observe these images in an SEM. In the near future the rudimentary grazing incidence mirror will be replaced by a diamond-turned ellipsoidal mirror of
much superior optical quality. We anticipate an increase of two orders of magnitude in the soft X-ray intensity at the environmental chamber which should enable the resist to be well exposed.

IV. MICROSCOPE DEVELOPMENT

In future work we plan to use the contact microscope to examine live specimens. The cells will be placed or grown on a suitable resist-coated substrate. This would be brought into contact with the window and exposed with the laser beam. Subsequently, the resist would be ultrasonically cleaned, developed, and examined either by phase or electron microscopy.

In addition, a new type of soft X-ray laser microscope has been constructed and will be installed on the soft X-
ray laser in the near future. Called COXRALM (composite optical X-ray laser microscope), this device is an inverted phase contrast microscope with the capability of observing UV-induced fluorescence combined with the option of contact micrograph generation via flash soft X-ray exposure. COXRALM, which is a collaborative effort by biologists and physicists, will provide the advantage of being able to observe the specimen until the time of X-ray exposure. This will directly address the question of specimen condition at exposure and permit the study of dynamic processes in cells.

V. PROGRESS TO SHORTER WAVELENGTHS

The sequence of Li-like ions represents an attractive avenue to progress to laser action at soft X-ray wavelengths shorter than those of current X-ray lasers [1]-[3]. Extension of the operating range of soft X-ray lasers to shorter wavelengths is important for applications such as microscopy of live biological specimens, and two approaches to the shorter wavelength region are under intensive development at Princeton. In this section we report on experiments to generate gain in lithium-like ions produced by a 1 kJ CO2 laser and confined in a magnetic field of up to 90 kG. The gain is generated at wavelengths of 15.4 nm in AlXII and 12.9 nm in SiXII. The approach is similar to our work on hydrogen-like carbon, CVI, which led to laser action on the n = 3 to m = 2 transition at 18.2 nm.

Gain measurements in Li-like ions in a recombining plasma have been reported in a series of experiments by Jaegle et al. [4]. In these experiments a Nd laser generates a plasma which expands freely, and a gain-length of 2-2.5 has been observed on the 5f-3d transition in AlXII at 10.6 nm. In parallel with the work at Princeton on CVI, early investigations of Li-like spectra from magnetically confined recombining plasmas showed evidence of population inversions [12], [13]. Time-integrated (photographic plate) and time-resolved line intensity measurements of the 4d-2p and 3d-2p transitions in CVI showed a 4d/3d population inversion. The 4f and 4d levels are expected to be collisionally populated in proportion to their statistical weights and the estimated 4f-3d gain is 1.8 cm\(^{-1}\) at 52 nm. A 4d/3d population inversion was also observed in Li-like NeVII in a gaseous target. An advantage of using lithium-like ions is that the ionization potential is lower than that for a hydrogenic-ion with a comparable lasing wavelength. For example, SiXII has an ionization potential of 523 eV for a 4f-3d wavelength of 12.9 nm. In comparison, hydrogen-like NVII has an ionization potential of 667 eV and a potential lasing transition (3 \(\rightarrow\) 2) at 13.4 nm. On the other hand, since the plasma electron density is limited to approximately 10\(^{20}\) cm\(^{-3}\) by the wavelength of the CO2 laser, the ion density of SiXII is necessarily lower than that of NVII. The trade-off between these and other factors can only be determined experimentally.

VI. EXPERIMENT WITH LI-LIKE IONS

The experimental setup is similar to that for CVI (shown in Fig. 1). The CO2 laser is focused onto an aluminum or silicon disk and creates a plasma of helium-like ions which is confined by a strong ( \(-50\) kG) magnetic field to a long, thin geometry suitable for high gain. The maximum laser power density on target is 2 \(\times\) 10\(^{13}\) Wcm\(^{-2}\). A composite blade made from a sandwich of aluminum and stainless steel (or silicon/titanium for silicon targets) is attached perpendicularly to the target surface (see Fig. 5). The blade helps to create a more uniform plasma in the axial direction and provides additional cooling by heat transport from the plasma to the blade and by additional radiation losses from the stainless steel or titanium. The plasma cools rapidly after the laser pulse and fast three-body recombination, followed by cascading processes, provides a high 4f population while the 3d level decays rapidly by the 3d \(\rightarrow\) 2p radiative transitions, and in this way a 4f-3d population inversion and gain is built up. Maximum gain occurs in the high-density, low-temperature off-axis regions, and a slot in the target disk transmits the axial stimulated emission. A second multichannel soft X-ray spectrometer [14] views the plasma transversely.

The axial emission is imaged by a grazing-incidence mirror onto the entrance slit of a multichannel soft X-ray spectrometer. The mirror is constructed by bending a glass strip; consequently, the optical quality of the system is not ideal. Hence a transverse scan of the axial spectrometer gives information on the relative divergence of the stimulated emission compared to spontaneous emission lines. In lithium-like ions the 4d-3p and 4p-3s transitions have gA values significantly less than the 4f-3d transition. (The upper-level statistical weight is g, and A is the transition probability.) In the presence of a n = 4, m = 3 population inversion, gain is expected to be first apparent in the transition with the highest gA value, i.e., 4f-3d, and one may expect to see its intensity increase on axis due to stimulated emission. A convenient measure of this increase is the 4f-3d intensity relative to the 4d-3p and 4p-3s transitions. This comparison, being based on lines in the same ion from upper levels with the same principal quantum number, is independent of uncertainties in the exact spatial distribution of the ions viewed by the
Fig. 6. Emission from the aluminum target assembly. (a) and (b) show the axial emission recorded with the grazing-incidence mirror installed. (a) is the on-axis emission and shows strong stimulated emission on the Al X\textsubscript{I} 15.4 \text{ nm} line. In (b), the axial instrument was moved transversely 200 \text{ pm} to record the off-axis emission. (c) and (d) show the transverse emission spectrum observed and that calculated in a computer simulation.

spectrometer. Fig. 6(a) and (b) shows the spectra recorded by the axial spectrometer at two different transverse positions. The ratio of the Al X\textsubscript{I} 15.4 \text{ nm} (4f-3d) to Al X\textsubscript{I} 14.1 \text{ nm} (4p-3s) line intensity is three times higher in the on-axis spectrum, Fig. 6(a), than in the off-axis spectrum recorded with the same instrument, Fig. 6(b). Further off-axis the ratio is higher, with a corresponding on-axis gain-length, \( gL = 3-4 \) (see Fig. 7(a)). The Al X\textsubscript{I} 15.4 \text{ nm} line in the transverse spectrum, Fig. 6(c), is blended with FeV\textsubscript{II} lines; however, the Al X\textsubscript{I} 5.24 \text{ nm} doublet is seen in third order with the components in a 2:1 intensity ratio, indicating that this high-density plasma is optically thin. This is important, since it permits the Al X\textsubscript{I} 5.24 \text{ nm} transition to efficiently depopulate the 3d level and generate a population inversion.

Similar experiments have been performed with SiX\textsubscript{II}. In this case the 4f-3d gain transition is at 12.9 \text{ nm} and the neighboring 4d-3p and 4p-3s lines are at 12.6 \text{ nm} and 11.9 \text{ nm}, respectively. The target configuration was similar to Fig. 5, but with a silicon disk and a silicon/titanium composite blade. The titanium provides additional radiation cooling. Since the SiX\textsubscript{II} ionization potential (523 eV) is higher than that of Al X\textsubscript{I} (442 eV) and the gain wavelength is shorter, it is expected to be more difficult to generate gain in SiX\textsubscript{II} than Al X\textsubscript{I}.

Fig. 7. (a) Observed and predicted line intensity ratio versus horizontal position of the axial spectrometer showing the rise in relative intensity of the Al X\textsubscript{I} 4f-3d transition at 154.7 \text{ A} in the region of the plasma with gain: experimental data, •: modeling with peak gain-length product, \( GL = 1.6 \), +: modeling with peak gain-length product, \( GL = 3.7 \). (b) The experimental line intensity ratios of SiX\textsubscript{II} 129 \text{ A} to 126 \text{ A} are shown as a function of the transverse position of the axial spectrometer.
Preliminary results are shown in Fig. 7(b), where the relative divergence of the 12.9 nm line is shown compared to the 12.6 nm and 11.9 nm lines. The peak in the intensity ratios is a clear indication of gain-length of order \( gL = 1-2 \), and we expect that with further optimization of experimental conditions higher values of gain will be obtained.

VII. Modeling of Lithium-Like Plasmas

A computer code [15] developed to simulate a laser-produced, hydrogen-like carbon plasma confined in a magnetic field has been adapted to model the above experiment with lithium-like ions [16]. The program consists of a hydrodynamic code and an atomic physics code [17]. The one-dimensional Lagrangian hydrodynamic code calculates, in cylindrical geometry, various time-dependent plasma parameters such as electron temperature, electron density, and the ground state populations of each ionization stage of the species considered (aluminum in the present case). These parameters are inputted in the atomic physics code which evaluates the populations of excited levels in AI XI, and the gain on the 4f-3d transition. It also generates radial profiles of the line intensities of various important transitions such as the 4f-3d, 4d-3p, 4p-3s, 4d-2p, 3d-2p, 3p-3s, and 3s-2p as a function of time. In the gain calculation, the line profile is assumed to be dominated by Doppler broadening, and the plasma length was taken to be 1 cm.

Excited levels up to \( n = 12 \), i.e., 77 levels, are included in the calculation. For conditions in which gain is predicted, i.e., low-electron temperature (10 eV) and intermediate electron density \(( \sim 5 \times 10^{18} \text{ cm}^{-3})\), excited levels above \( n = 5 \) are expected to be in LTE with the ground state of the He-like ion, Al XII. Hence the populations of excited levels from \( n = 6 \) to 12 are calculated from the Saha–Boltzmann equation. The populations of other states are calculated from a collisional–radiative model, i.e., all radiative decay rates, collisional excitation and de-excitation rates between levels, and collisional ionization rates from and radiative and three-body recombination rates to each level are included.

Fig. 8 shows electron temperature, electron density, and gain profiles at the time when the gain reaches its maximum in Al XI. The laser pulse is 50 ns FWHM, and 7 percent of iron is included to provide additional cooling. The gain peaks at 87 ns and has a duration of about 20 ns in this particular run (the laser pulse starts at \( t = 0 \)). The center of the plasma is heated directly by laser and a shock wave is generated which transports ions away from the central hot region, and produces favorable conditions for a gain of 1–1.5 mm off-axis. This is consistent with experimental observations [2].

To model the experiment, axial and transverse emission spectra are constructed by integrating the emission lines over time, including, in the axial spectrum, stimulated emission due to the presence of gain. The spectra are spatially integrated over the observation regions of the spectrometers in the experiment. One can see in Fig. 6(c) and (d) that the predicted relative intensities of the spontaneous emission from the 14.1 nm (4p–3s), 15 nm (4d–3p), and 15.4 nm (4f–3d) transitions agree well with the observed transverse spectrum.

As explained above, the experimental data in Fig. 7 represent both spatial and angular information on the line intensity ratios. It was possible to simulate the spatial variation of relative line intensity by spatially integrating over different plasma regions corresponding to the different regions viewed by the axial spectrometer during the transverse scan. This was done for two model plasmas with peak gain-lengths \( gL = 1.6 \) and 3.7, respectively. These different values of gain-length were generated by changing the amount of additional cooling material for a constant laser input energy (7 percent of iron for \( gL = 3.7 \) and 5 percent of iron for \( gL = 1.6 \)). The results are shown in Fig. 7(a), together with the experimental data. It can be seen that there is overall good agreement between the shape of the curves for \( gL = 3.7 \) and the experimental data. The similarity in the shape of the curves is consistent with an experimental gain value of \( gL = 3-4 \).

VIII. Cavity Development

We are also working on cavity development with the aim of increasing the brightness of our soft X-ray laser. Without mirrors, the laser beam divergence is governed solely by the plasma geometry—any ray that can pass through the gain region can be amplified and the divergence at present is 5–10 mrad. With a properly designed laser cavity the divergence could be near diffraction limited with a potential increase in brightness of up to \( 10^6 \). To properly establish the cavity modes, however, many passes through the gain medium are needed and a long duration gain is necessary. The Princeton 18.2 nm laser with a gain duration of 10–30 ns is ideally suited to cavity
development.

Early work at Princeton using newly developed multilayer mirrors in a double-pass arrangement resulted in a factor of two intensity increase due to the amplification of the stimulated emission in the second pass [18]. Mirror alignment posed severe difficulties; however, a new experimental arrangement designed to overcome these problems has been constructed and cavity experiments are planned for the near future.

IX. TWO-LASER APPROACH TO WAVELENGTHS SIGNIFICANTLY BELOW 10 NM

The advantage of illumination in the 2.4–4.4 nm wavelength region is well known in microscopy. Extension of the operating range of current soft X-ray lasers to the 2.4–4.4 nm range is a significant technical challenge due to the severe demands placed on pump laser power at shorter wavelengths (e.g., [19]). The equipment cost may be reduced, however, by using a very short pulse laser. A picosecond laser pulse length is also well matched to the radiative time scales in candidate ions.

An experiment to investigate a two-laser approach to laser action at wavelengths significantly below 10 nm has been conducted at Princeton with the first experiments conducted in the fall of 1987. The basic approach is to split the task of creating a soft X-ray gain between two pump lasers. First, a CO2 or Nd laser creates a highly ionized plasma containing ions of the appropriate ionization stage confined in a magnetic field. Then a powerful picosecond laser ($I \sim 10^{18} \text{ W/cm}^2$) populates a selected excited state in the ion by multiphoton excitation. At these laser powers very high-order multiphoton transitions are efficiently excited and are expected to selectively populate a specific state, generating a population inversion. Candidates include inner shell and doubly excited transitions in argon-like and krypton-like ions [20]. This approach was stimulated by the observation of stimulated emission at 931 Å by multiphoton excitation of an inner-shell transition in neutral krypton [21]. Other schemes, including one based on inner-shell ionization, are also under active consideration.

The first stage in the construction of the experimental system is now complete. The powerful picosecond laser now produces 20–25 mJ at 248 nm in a 1.1 ps pulse. The target chamber, magnet system, and diagnostics and control systems are operational and we have recorded the first plasma spectra produced by the combined system, including the CO2 laser. The spectra showed a dramatic increase in line emission in the presence of the magnetic field.

While work was in progress in the preparation of the complete system, high-resolution soft X-ray spectra were obtained from the powerful picosecond laser alone. We have identified emission lines from ionization stages as high as Fe$^{5+}$, a result which illustrates the strength of the laser-target interaction at ultrahigh intensities, $I > 10^{16} \text{ W/cm}^2$ (see Fig. 9). Very broad spectral line profiles of FVII were also recorded. Line profiles can be a sensitive indication of plasma conditions, and the appearance of the forbidden 2p–3p component in the FVII 2p–3d transition was correlated with theoretical Stark-broadened profiles at electron densities $N_e = 10^{17} \text{ cm}^{-3}$ [22].

We have obtained laser action at 248 nm in a large aperture (7 cm × 10 cm) KrF amplifier under development at Princeton. This discharge pumped device will be used to increase the output energy of the powerful picosecond laser to the joule level corresponding to an intensity on target in excess of $10^{18} \text{ W/cm}^2$. At this level the electric field due to the laser is much higher than the Coulomb field between the electrons and the nucleus, opening up a new area of physics with applications not only to X-ray lasers but also to chemistry and biology.

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References


[16] D. Kim et al., "Soft X-ray amplification in lithium-like AlXII (154 Å) and SiXII (129 Å)," submitted to J. Opt. Soc. Amer. B.


