Soft X-ray Lasers and Their Applications

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The emerging technology of soft x-ray lasers has novel applications to microscopy, lithography, and other fields. This article describes the status of soft x-ray laser research with the aim of bringing the rapid developments in this field to the attention of potential users in other disciplines. The different techniques for generating a population inversion and producing a soft x-ray laser are reviewed. The status of current research in the field and the near-term prospects are described. It is expected that the range of potential applications of soft x-ray lasers will increase as their performance improves. Work aimed at increasing the output power and progressing to shorter wavelengths with these devices is also reviewed.

ASERS OPERATING AT X-RAY WAVELENGTHS HAVE BEEN sought after since the invention of the first lasers in 1960, because of their intrinsic interest and also because their ultrahigh brightness and short wavelengths made them ideal for a variety of applications. However, the creation of a population inversion in highly ionized ions necessary for x-ray laser action places severe demands on the required pump source, and for this reason soft x-ray lasers remained an elusive dream until 1984. The first demonstration of lasing action in the soft x-ray region by groups at Lawrence Livermore National Laboratory and Princeton University in 1984 has been followed by work at a number of laboratories around the world. At present, rapid progress is being made in the extension of the operating wavelength range and power of these devices and in the development of small-scale and relatively low-cost soft x-ray lasers suitable for widespread application to fields such as microscopy and microelectronics.

Recent work at a number of laboratories has been aimed toward the development of soft x-ray lasers in wavelength regions shorter than 20 nm and of lasers in the vacuum-ultraviolet (VUV) region near 100 nm and below. Noteworthy are the achievements of very high gain (much above saturation) in Cs near 90 nm with relatively low pumping power as demonstrated by the Stanford group (1), the generation of very high power density radiation (above 10^{18} W cm⁻²) for application to soft x-ray lasers by the Chicago group (2), and theoretical work on "tabletop" VUV lasers at 30 to 40 nm with the use of Ni-like or Nd-like ions by the Massachusetts Institute of Technology group (3). The experimental and theoretical work at Princeton (4–8) includes: (i) the development of a powerful subpicosecond 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 electromagnetic fields; and (iii) improvement of the present 18.2-nm laser and its application to soft x-ray microscopy. The existing soft x-ray laser at Princeton is pumped by a commercial 300-J CO₂ laser and has a wavelength of 18.2 nm, an output energy of 1 to 3 mJ, a pulse duration of 10 to 30 ns, and a beam divergence of 5 mrad.

Soft X-ray Laser Research

Although soft x-ray laser research was pursued in the 1970s, the field came of age in 1984 with the first unambiguous demonstration of high gain by groups at Lawrence Livermore National Laboratory (9) and Princeton University (10). These two groups used different approaches to generate gain (see Fig. 1), and these approaches have formed the basis of all successful work on soft x-ray lasers since that time. (i) The recombination approach is based on H-like or Li-like ions. (ii) The collisional excitation approach is based on Ne-like or Ni-like ions.

Both schemes rely on high-power pulsed lasers to create the appropriate conditions in a plasma, and in both schemes the population inversion necessary for stimulated emission and gain is brought about by fast radiative decay of the lower level. In the recombination approach for H-like ions, a laser is used to create a plasma with a high fraction of totally stripped ions. After the laser pulse, the plasma is cooled rapidly and undergoes fast three-body recombination. In some cases the plasma is cooled by adiabatic expansion. One unique feature of the Princeton laser is that the plasma is confined in a magnetic field and cooled by radiation losses. The magnetic field maintains a high electron density, which is beneficial because the three-body recombination rate scales as the electron density squared. It also helps to shape the plasma into a long thin geometry suitable for a laser and enables an efficiency to be attained that is almost 100 times higher than other operational soft x-ray laser systems. Three-body recombination puts into upper excited levels a high population, which decays downward by collisional radiative cascade. In hydrogenic ions, level 2 decays rapidly by radiation and a population inversion is built up between levels 3 and 2. The atomic structure of Li-like ions is similar to that of H-like ions, and the same approach works in this system also (11, 12). In this case the 3-2 transitions have a high radiative decay rate, and gain can be generated on the 4-3 and 5-3 transitions. The Lilike sequence has the advantage of a shorter wavelength lasing transition for ions of similar ionization potential, that is, a better 'quantum efficiency."

The Ne-like scheme was applied at Lawrence Livermore National Laboratory. Here a high-density, high-temperature plasma is generated by a large Nd laser, Novette or Nova. 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

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the 3p and 3s levels. The same scheme also works in Ni-like ions, and here, as in the case of Li-like ions, there is an advantage in using the Ni-like sequence to access the shortest possible wavelengths. A recent review of work at Livermore is given in (13). The ambitious goal of the Livermore work is to develop a high-brightness, highcoherence soft x-ray laser in the wavelength range 4.0 to 5.0 nm for high-resolution holographic imaging of biological specimens. One step in this direction was the recent achievement of a gain-length (GL) of \approx 4 at 4.48 nm in Ni-like Ta. In summary, both approaches use a laser to create an appropriate plasma and rely on fast radiative decay to deplete the lower level in order to generate a population inversion. The major difference is that in one case the upper level is populated through recombination and in the other it is populated by collisional excitation.

A number of laboratories around the world are heavily engaged in soft x-ray laser research. We would like to mention the pioneering work of Pert and Ramsden and their colleagues (14) at Hull in England on the recombination scheme for C fibers at 18.2 nm, which was later taken up at the Rutherford Appleton Laboratory (15). This work is now part of an international effort involving seven different institutions in England, France (Orsay), Japan (Institute for Laser Engineering), and the United States (Naval Research Laboratory). The soft x-ray laser experiments of Jaeglé et al. (11) originated from the observation of a bright line at 10.57 nm in an Al plasma. This previously unknown line was identified as the 3d-5fline of Li-like Al. Confirmation that this line was due to amplification of spontaneous emission was obtained from measurements of the plasma absorption or gain over the observed spectrum. A clear peak was seen at 10.57 nm, corresponding to a gain of up to 2.5 cm⁻¹. More recently, experiments with a 6-cm-long plasma have resulted in a gain-length of $GL \approx 3.0$. Use of a soft x-ray mirror with a reflectivity of 5% in a double-pass arrangement resulted in an effective gain-length of $GL \approx 4$. Gain-lengths of up to $GL \approx 2.7$ on the 3d-4f and 3d-5f transitions in Li-like Al have also been reported



Fig. 2. Plot of gainlength achieved to date versus wavelength (5-20). The dashed line marks GL = 4.6, which corresponds to an enhancement of 10. by Moreno et al. (16).

The recombination approach has been used at the Laboratory for Laser Energetics at Rochester University (17) to produce gain on the CVI 18.2-nm transition in a radiation-cooled Se/Formvar plasma. The collisional excitation scheme has also been demonstrated by a group at the Naval Research Laboratory (18) at wavelengths from 19.5 to 28.5 nm in Ne-like Ge and Cu. This experiment used less demanding technology than the scheme at Livermore: a lower power driver laser (350 to 485 J operating at the fundamental wavelength of $1.05 \mu m$) and simple slab targets rather than exploding foils. Interestingly, the J=2-J=1 gain coefficient (J is the angular momentum quantum number) of $G = 4.1 \text{ cm}^{-1}$ observed with single-sided illumination of a thick slab of Ge was comparable to that obtained by the Livermore group with an Se film target that was illuminated from both sides. Also for the first time the Cu J=0to J=1 line showed a G comparable to that for the J=2-J=1 lines, in agreement with theoretical predictions. In Japan, Herman et al. (19) have demonstrated gain at various wavelengths including a measurement of G of 2 cm⁻¹ in the 8.1-nm transition in H-like F. Kato et al. (20) have reported GL = 1 at 5.4 nm and GL = 0.9 at 4.5 nm, but their experimental data have been criticized as being ambiguous and unconvincing. Very recently there has been a report by Hara et al. (21) of gain in the soft x-ray region produced by a small-scale (6-J) pump laser. The presence of gain was deduced from the nonlinear rise in intensity with length of Al X and Al XI emission lines. However, in contrast to earlier work by Kim et al. (5, 6, 12), the nonlinear rise of the "gain lines" with plasma length was not referenced to a linear rise of "no-gain lines" in Al XI such as the 14.1-nm transition, leaving open the possible influence of other effects (such as gradients in the plasma temperature along the plasma region viewed by the detector) that could also produce a nonlinear intensity rise. Clarification of this point would be valuable as this result, if verified, would add to earlier work (5, 6) with very favorable implications for the commercialization of soft x-ray lasers.



A third potential method to achieve soft x-ray lasing is resonant

Fig. 3. Carbon spectra obtained with a Teflon target (**A**) without pulse compression [20-GW laser power; E = 20 mJ (200 shots), $\Delta t = 1$ ps] and (**B**) with pulse compression and final KrF* amplifier [0.3-TW laser power, E = 100 mJ (5 shots), $\Delta t = 300$ fs]. [Adapted from (4) with permission of the Optical Society of America]

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photopumping in a two-component plasma (22). Resonant photoexcitation has been recently demonstrated in the soft x-ray range by Monier *et al.* (23). A H-like Al XIII resonance line was used to pump a Ne-like Sr XXIX resonance line, resulting in an increase of a factor of 2 in fluorescence from the upper state of the pumped transition. However, the pumping efficiency was less than expected. Several difficulties need to be overcome if this method is to be successfully applied to generating soft x-ray lasing.

The current state of the art is represented in Fig. 2, which shows GL versus wavlength. High GL values have been achieved over a variety of wavelengths, although in general there is a falloff in gain as the wavelength approaches the "water window" region, 2.4 to 4.4 nm, important for biological applications. It is only when GL exceeds 4.6 that the output intensity exceeds spontaneous emission by more than an order of magnitude and one can talk about a soft x-ray laser beam. A number of groups are investigating the physics relevant to more efficient or shorter wavelength schemes, in particular Harris and his colleagues (24) and Murname *et al.* (25), who are working on Auger ionization, and Boyer *et al.* (26), who are working on multiphoton processes. More details of the current research directions are contained in (27). For special journal issues devoted to soft x-ray lasers, see (28, 29).

Soft X-ray Laser Research at Princeton

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 soft 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 (30)]:

where

$$\Delta N_{\rm inv} \sim P$$

Therefore, in order to decrease the lasing wavelength from 10 to 1 nm, the pumping power must be increased approximately by a factor of 10^4 .

The recombination laser at Princeton 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 need to be on the order of tens of megajoules. 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 soft x-ray laser development is being devoted to schemes in which metastable and autoionizing levels can be used for storing pumped energy, as proposed by Harris (31), or schemes based on very short (picosecond and subpicosecond) pumping pulses (32). This second approach is particularly attractive because the upper state lifetime is of the order of 10^{-12} to 10^{-13} s for a transition wavelength of ≈ 1 nm. Therefore, pumping is required only for a picosecond or subpicosecond time interval. 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, $P \sim 10^{12}$ W. Even more important, such a laser operating in the UV range (for example, the KrF excimer laser with wavelength 0.25 µm) can be focused to a 2to 3-µm spot, providing a power density in excess of 10^{18} W cm⁻² (with corresponding electric field $\sim 10^{11}$ V cm⁻¹ = 10 kV nm⁻¹) on target. With this power density it is possible to provide multiphoton excitation and multiphoton ionization of highly ion-

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ized 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, we proposed in 1986 the use of two lasers (33). The role of the first, high energy but low-power laser (for example, 0.5kJ, 50-ns CO₂ or 100-J, 3-ns Nd/YLF laser, where YLF is yttrium lanthanum fluoride) is to create a plasma column of highly ionized ions that may be confined in a strong magnetic field. The role of the second, extremely high power laser (~1-TW KrF laser) is to generate gain by multiphoton ionization, very fast ionization (for example, inner shell ionization), or selective multiphoton excitation. More details about the Powerful Picosecond (Sub-Picosecond) Laser (PP-Laser) system are presented by Meixler *et al.* (4).

One of our first experiments with such a high-power density laser beam was a measurement of soft x-ray spectra for C and F. The laser beam was focused on a rotating cylindrical Teflon target while a soft x-ray, grazing incidence Schwob-Fraenkel spectrometer (SOXMOS) with multichannel detector monitored the plasma radiation from the target surface. Figure 3B shows the spectrum in the vicinity of the C VI 3.37-nm and C V 4.03-nm lines (both from $2 \rightarrow 1$ transitions). In addition to the enormous line broadening, one can see a strongly pronounced, unusual structure in the lines. Both broadening and satellite structure are larger in the spectrum obtained at high power than in the spectrum obtained at lower power (Fig. 3A). On the red side of the lines, several satellite lines can be identified (2p2s-1s2s at 3.42 and 3.45 nm; 2s²-1s2p at 3.55 nm; and 1s2s2p-1s²2s at 4.09 nm). In addition, there is a component at 3.3 nm on the blue side of the CVI 3.37-nm line. It should also be noticed that the number of shots needed for these short-wavelength spectra is proportional not to the laser beam energy but rather to its power. (The energy of the laser beam increased by a factor of 5 while the number of shots decreased by a factor of 20.) In spectra obtained earlier with the 20- to 30-GW PP-Laser (10¹⁶ W cm⁻²), part of the large broadening and asymmetry of the F VII lines in the spectral region from 12.0 to 14.0 nm was attributed to the Stark effect and radiation from the forbidden components of the lines (34). Very recently, Koshelev (35) interpreted asymmetric broadening of the F VII lines to be a result of satellite line radiation. Spectral lines of C VI, C V, and F VII 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 partially responsible for these effects. Spectroscopic measurements 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.

Development of an additional amplifier at 18.2 nm. Presently, the highest beam energy of our soft x-ray laser (SXL) (36) at 18.2 nm (C VI 3-2 transition) pumped by a 300-J CO₂ laser in a 90-kG solenoidal magnetic field is 3 mJ with a 3-min repetition rate. At present, the beam energy is one of the most important parameters for applications of the SXL. In order to increase it, we have developed an additional SXL amplifier (3 mm long) at 18.2 nm that is pumped by a Nd/YLF laser beam line-focused onto a C target. Thin Fe 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 15 J of Nd/YLF laser beam energy on target [see Kim *et al.* (5, 6)]. The axial spectra in the vicinity of the lasing line C VI 18.2 nm (3-2 transition) and in the vicinity of C VI 13.5 nm (4-2 transition) were measured for 1-, 2-, and 3-mm-long targets. A striking, nonlinear increase in the intensity of the 18.2-nm line, in comparison to near-linear increase in intensity of the 13.5-nm line, may be seen in Fig. 4.

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



Fig. 4. Gain measurement for C VI 18.2 nm with the use of 15-J pump laser energy on target (25 J in total beam; 50 kG, stainless steel blade). [Adapted from (6) with permission of the Optical Society of America]

Cavity development. A laser cavity can increase the brightness of the SXL beam by several orders of magnitude by decreasing the divergence to a value 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 (GL > 4 for several cavity round-trip times). In our early work, using a newly developed multilayer mirror (37), we demonstrated a 120% increase in 18.2-nm radiation, due to amplification of stimulated emission, by using a mirror with a reflectivity of only 12% (38). However, the mirror alignment posed tremendous difficulties, which practically made it impossible to develop a cavity in the original SXL setup. We have therefore designed an unstable resonator-type cavity with a transversely pumped C fiber as the lasing medium. The same cavity will also be used for a 1-cm-long SXL created by a line-focused Nd/YLF laser beam incident on the cylindrical C target described in the section on an additional amplifier.

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 soft x-ray radiation. Recently, however, Ceglio *et al.* (39) 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.

Gain in Li-like ions at 15.4 and 12.9 nm. Pioneering work for Li-like Al XI ions, particularly for the 5*f*-3*d* transition at 10.5 nm, was done by Jaeglé and his group (11), using a Nd/glass laser (initial plasma electron density $N_e = 10^{21}$ cm⁻³) for the pumping lasing medium.



Fig. 5. Schematic of the COXRALM designed to allow biologists to select and observe live cells before recording a high-resolution contact image with the soft x-ray laser.

In our system with a CO₂ pump laser the initial electron density is $N_e = 10^{19}$ cm⁻³. For such an electron density, the largest gain in Al XI and Si XII is expected for the 4*f*-3*d* transitions at 15.4 nm and 12.9 nm, respectively. The Al or Si targets used in the experiment were very similar to the SXL C 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 $GL \approx 3$ to 4 for 15.4-nm radiation and $GL \approx 1$ to 2 for 12.9-nm radiation. Details about the experiment and theoretical modeling are presented in (5, 12).

Application of the Soft X-ray Laser to Microscopy

Most of what is known about the internal structure of cells has been learned by the development and application of the techniques of electron microscopy. This knowledge rests on the premise that the intensive procedures necessary to prepare a specimen for electron microscopy do not significantly influence the structure, form, and high-resolution detail observed. Nonetheless, unanswered questions remain about the fidelity of the image of a cell that has been fixed, stained with heavy metals, and sectioned to the original living cell. X-ray microscopy offers a new way to look at unaltered cells in their natural state. The absorption edges in the x-ray spectra of naturally occurring cell constituents provide contrast without the addition of heavy metals to the cell necessary in electron microscopy. Work has begun in using high-brightness synchrotrons and soft xray lasers as light sources for x-ray microscopy (40). Biologists have long dreamed of observing the form and function of living cells at high resolution. The short, nanosecond pulse length of soft x-ray lasers offers the potential of observing a cell that was alive the instant before a flash exposure of a soft x-ray laser recorded its image. The necessary radiation dose levels make it unlikely that the cell will survive the exposure, but exposures of different cells should make it possible to piece together new information about dynamic processes inside cells.

Work has begun at Princeton to use the 18.2-nm laser (SXL) for soft x-ray contact microscopy of biological specimens. (This work is also closely related to x-ray microlithography.) The ultimate goal of our x-ray laser microscopy program is to obtain images of living cells. The details of this work as well as our other work with soft xray laser microscopy are described in (7, 41, 42).

In the x-ray laser microscope, a thin (~ 0.1 -µm) silicon nitride window square separates the vacuum tube, in which x-rays travel, from the biological cells located on photoresist at atmospheric pressure. We have demonstrated that the SXL beam has sufficient energy to expose images on photoresist in a single shot. In x-ray contact microscopy, magnification is obtained when the exposed photoresist is viewed in a scanning electron microscope (SEM). Images of diatom fragments (the silicified skeleton of planktonic algae) on photoresist indicated that the resolution on the photoresist was better than 0.1 µm. One may also regard diatom fragments as a kind of lithographic mask and an illustration of the potential application of the SXL to microlithography.

We have built a Composite Optical/X-ray Laser Microscope (COXRALM), shown schematically in Fig. 5 and described in (7, 42). COXRALM is designed to allow a biologist to select and observe live biological cells, using an optical phase-contrast microscope (43), and then create a high-resolution image of the cells on photoresist with the SXL beam. The first results were obtained with dehydrated cells to optimize image contrast and resolution without the technicalities of handling wet, live cells. Figure 6 shows a SEM image of a replica produced by the 18.2-nm SXL of dehydrated



Fig. 6. False-color SEM image of a replica of a HeLa cell that was subject to a viral infection. The replica was generated by contact microscopy with the 18.2-nm laser. The computer-generated false color enhances the contrast of the cell features. Scale bar, 10 µm.

HeLa cells (Helen Lane cervical cancer cells) obtained from the Biology Department of Princeton University. Presently our work is concentrated on experiments with live cells in a wet environment and on the development of a new Imaging Soft X-ray Laser Microscope (IXRALM).

Future Prospects

Rapid progress in soft x-ray laser development has been made. Especially exciting is progress in the miniaturization of soft x-ray lasers and the start of work on their applications. The general impact soft x-ray lasers will have in science and technology will depend on improvements in both their performance and their cost. It is necessary for their successful commercialization that these devices operate routinely at high gain-lengths (GL > 4), with the use of a low-cost driver laser, and this needs more system development and engineering. Most applications of visible-wavelength lasers are based on the fact that the brightness of these lasers is several orders of magnitude greater than that of conventional spontaneous emission sources, and this is achieved principally by the laser cavity mirrors. This technology is significantly more difficult in the x-ray region because of intrinsic limitations of x-ray absorption in materials and present limits in the soft x-ray laser pulse lengths. Nonetheless, a "revolution" (44) in x-ray optics is under way and the precedent of visible-wavelength lasers illustrates the potential benefits awaiting the creative inventor of applications of this technology to novel fields.

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