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X-Ray Lasers

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X-Ray Lasers

Jorge J. Rocca

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49.1 Introduction

Specific proposals for excitation schemes for X-ray lasers date back to 1967, when the possibility of achieving amplification by photoionization pumping was first suggested by Duguay and Rentzepis [1]. This was followed by proposals for X-ray lasers based on the two population inversion mechanisms on which the present day’s soft X-ray lasers are based: electron impact excitation schemes [2–5] and collisional recombination [6–9].

Several experiments realized during the 1970s and early 1980s yielded the observation of population inversions and gain [10–16]. Nevertheless, the experimental demonstration of large amplification at soft X-ray wavelengths was not realized until 1984, when Matthews et al. [17] and Suckewer et al. [18] observed amplification from the generation of population inversions in plasmas by collisional electron excitation and collisional electron–ion recombination, respectively. Subsequent experiments greatly expanded the number of laser transitions, achieved soft X-ray laser operation in the saturated regime for the first time [19–25] and realized proof-of-principle demonstrations in several applications [26–30]. However, the large size, cost and low-repetition rate of these lasers were barriers to their widespread utilization. Much progress has been achieved in the development of compact soft X-ray lasers. This is the result of advances in pump sources that include the development of compact multi-terawatt ultrashort pulse optical laser systems based on chirped pulse amplification [31–35], and fast capillary discharges capable of generating highly ionized plasma columns with high axial uniformity and length-to-diameter aspect ratios of 1000 to 1 [36–46]. Also contributing to the progress towards practical tabletop
X-ray lasers is the implementation of excitation mechanisms that take full advantage of the high intensity and ultrashort pulsewidth of new optical laser systems. The field is rapidly approaching the stage at which soft X-ray lasers sufficiently compact to fit onto a normal optical table (frequently described as ‘tabletop’ lasers) will be routinely utilized in science and technology.

Several articles have reviewed the progress in X-ray laser development [47–52]. The book by Elton discusses the developments up to 1990 [53]. A detailed account of the progress realized since that time1 can be found in the proceedings of two biannual conferences that focus on soft X-ray lasers [54,55]. The advances in tabletop lasers have also been reviewed [56].

### 49.2 Amplification and Pump

#### Power Requirements

In contrast to most optical lasers, the duration of the gain in soft X-ray lasers is usually shorter than the time required for the effective use of an optical cavity. This is the result of either the rapid self-terminating nature of the population inversion in transient schemes [57–66] or the difficulty in maintaining the stringent plasma conditions necessary for amplification in quasi-cw schemes for a sufficient period of time. Consequently, soft X-ray lasers normally operate with single- or double-pass amplification of the spontaneous emission through the gain medium. In such amplifiers, \( I \), the spectrally integrated intensity of the laser line, increases as a function of \( l \), the plasma length, as

\[
I = \frac{E g}{g} (e^{gl} - 1)^{1/2} g (e^{gl})^{-1/2}
\]

where \( g \) is the small-signal gain coefficient (assumed to be constant along the plasma column) and \( E \) is a constant proportional to the emissivity [67]. The product \( gl \) of the small-signal gain coefficient and the length of the amplifier (known as ‘the gain–length product’) have been widely used in the literature as a figure of merit to describe the level of amplification achieved.

A gain–length product greater than 5, commonly measured by monitoring the growth of the laser line intensity as a function of plasma length, is usually a clear indication that lasing is taking place. However, there are, in the literature, quite a large number of X-ray amplification experiments that measured products less than 4. In these cases, the data should be treated with significant caution, as such, phenomena such as for example plasma column end effects, could be responsible for the apparent enhancement of the line intensity. Ideally, the nearly exponential amplification continues until the intensity approaches the saturation intensity, as illustrated in Figure 49.1, for a 46.9 nm Ne-like Ar capillary-discharge-pumped laser [37]. At this intensity, the stimulated emission is sufficiently strong to extract the majority of the energy stored in the population inversion, depleting the population inversion and the gain. In collisionally excited lasers, gain saturation typically occurs [21–24,63] with \( gI \approx 14–20 \). Consequently, to achieve saturated soft X-ray amplification in a single- or double-pass amplifier, the gain coefficient must be, depending on the length of the plasma, one to three orders of magnitude larger than those typically encountered in visible gas lasers which use optical cavities [68]. Moreover, the physics of the generation of amplification by stimulated emission determines a dramatic upward scaling of the power density deposition required to obtain substantial gain at ultrashort wavelengths. Consider a transition at wavelength \( \lambda \) from an upper level of degeneracy \( g_2 \) and population \( N_2 \) to a lower level of population \( N_1 \) and degeneracy \( g_1 \). The small-signal gain coefficient is the product of \( \sigma \), the stimulated emission cross section and the population inversion density, \( N^* \):

\[
N^* = \frac{N_2 - \frac{g_2}{g_1} N_1}{g_1}
\]

and

\[
g = \sigma N^* \approx \frac{A_{21} \lambda^2}{8\pi \Delta \nu} \left( N_2 - \frac{g_2}{g_1} N_1 \right)
\]

in which \( A_{21} \) is the Einstein coefficient for spontaneous emission and \( \Delta \nu \) is the linewidth of the transition.

Now, \( A_{21} \) scales along an isoelectronic sequence as \( A_{21} \propto \lambda^{-2} \). Considering, as an example, the case of a naturally broadened unbranched transition with linewidth \( \Delta \nu \approx A_{21} \), we obtain:

\[
g \propto N \lambda^2.
\]

In turn, \( P_{\text{min}} \), the minimum pump power density required to maintain a certain upper laser level population density \( N_2 \), scales as

\[
P_{\text{min}} = N_2 A_{21} \frac{hc}{\lambda}.
\]
It results from the two previous equations that the power density required to obtain a certain gain coefficient in a naturally broadened transition scales as

$$P_{\text{min}} \propto g \lambda^{-5} \quad (49.6)$$

Consideration of the case of Doppler broadening, the line-broadening mechanism most frequently dominant under soft X-ray laser operating conditions, leads to a different but also dramatic scaling of the power requirement. In this case,

$$P_{\text{min}} \propto g \lambda^{-4} \sqrt{\frac{k_{\text{B}} T_i}{M_i}} \quad (49.7)$$

in which $T_i$ is the kinetic temperature of the emitting species which has atomic mass $M_i$.

Nevertheless, using the case of natural broadening for purposes of illustration, the operation of a saturated, mirrorless laser at 50 nm would require an estimated pump power density $10^7$ times larger than that necessary for a 500 nm blue-green laser, while one operated at 5 nm would require an estimated pump power density $10^{12}$ times larger than that for the blue-green. In making these estimates, it is assumed that the use of the cavity for the 500 nm laser reduces the gain requirement for lasing by a factor of 100. In addition, the pump power required for lasing at soft X-ray wavelengths is often further increased by refraction losses that reduce the effective gain coefficient.

### 49.3 Soft X-Ray Laser Characteristics

#### 49.3.1 Wavelength

To date, saturated soft X-ray lasers have been developed at wavelengths ranging from 5.8 [21] to 46.9 nm [37,44]. Significant amplification has been observed at wavelengths as short as 3.56 nm [20].

#### 49.3.2 Output Pulse Energy

The output pulse energy generated by soft X-ray lasers exceeds, by several orders of magnitude, that of other sources of coherent short wavelength radiation. Saturated collisional lasers pumped by large optical lasers have achieved energies of several mJ (e.g. 8 mJ at 15.5 nm [24]), and an 18.2 nm recombination laser has been reported to emit 3 mJ pulses [18]. A highly saturated 46.9 nm tabletop discharge pumped laser has produced millijoule-level pulses at a repetition rate of 4 Hz [45]. Transient collisional lasers pumped by relatively compact optical lasers have produced picosecond laser pulses with energies of the order of 10 μJ [64].

#### 49.3.3 Pulse Duration

The pulsewidth of presently available soft X-ray lasers ranges from a fraction of a nanosecond to several nanoseconds for lasers operating in a quasi-steady-state regime, to several picoseconds for transient collisional excitation lasers [59–66] and recombination lasers in transitions to the ground state [57,58]. Future photoionization X-ray lasers will generate femtosecond pulses.

#### 49.3.4 Peak Power

Quasi-cw collisional electron excitation lasers have produced peak output powers of several tens of MW [69], and recombination lasers have reached peak powers of the order of 0.1 MW [18]. Very compact capillary discharge tabletop collisional lasers have reached peak powers up to 0.6 MW [45], and transient collisional lasers have generated powers of up to ~1 MW [64].

#### 49.3.5 Average Power

Most soft X-ray lasers demonstrated to date have produced low-average power due to the low-repetition rate used in the majority of the experiments that have produced significant output pulse energies (typically less than one shot every few minutes). An exception is a 4 Hz tabletop capillary discharge laser operating 46.9 nm that produced an average power of 3.5 mW [45]. The average power of soft X-ray lasers can be expected to increase dramatically during the next decade as a result of the development of efficient high-repetition-rate tabletop pump sources.

#### 49.3.6 Beam Divergence

In the presently available soft X-ray lasers, the beam divergence is dominantly determined by refraction and typically ranges from 1 to 10 mrad. Refraction bends the amplified X-rays out of the gain volume as a result of the transverse variation of the index of refraction due to the large electron density gradients.

#### 49.3.7 Temporal Coherence

Since presently available soft X-ray lasers do not employ optical resonators, their temporal coherence depends on the atomic linewidth of the laser transition. In most of these single- or double-pass amplifiers, the dominant line-broadening mechanism is the Doppler effect. Taking into account the fact that amplification narrows the output by a factor of $(gl)^{-1/2}$ below that of spontaneous emission from a thin source, the value of the linewidth $\Delta \nu_{\text{FWHM}}$ can be estimated as

$$\Delta \nu_{\text{FWHM}} = v_0 \sqrt{\frac{8k_{\text{B}} T_i \ln 2}{M_i c^2}} \frac{1}{\sqrt{gl}}. \quad (49.8)$$

For values of $gl$ above saturation ($gl \approx 14–20$ in collisional lasers), some line re-broadening may occur. This linewidth determines $l_{coh}$, the longitudinal coherence length, and $\tau_{coh}$, the coherence time, via

$$l_{coh} = c \tau_{coh} = \frac{c}{\Delta \nu_{\text{FWHM}}} \sqrt{\frac{2 \ln 2}{\pi}} = 0.66c \Delta \nu_{\text{FWHM}}. \quad (49.9)$$
A few measurements of the linewidth of saturated collisional excitation lasers have been reported [70,71] and correspond to $l_{\text{coh}} \approx 0.1–0.2 \text{mm}$.

49.3.8 Spatial Coherence

Several authors [72–74] have theoretically analysed the spatial coherence of soft X-ray amplifiers. Single-pass soft X-ray amplifiers can sustain a large number of spatial modes and, therefore, have poor spatial coherence [75,76]. However, significant improvements can be obtained by increasing the length of the plasma column [77] or by operating in a double-pass amplification [78]. The spatial coherence of a capillary discharge laser was measured to increase monotonically with plasma column length to reach nearly fully spatial coherence for plasma columns 36 cm in length [79]. This is the result of a decreasing number of spatial modes along the plasma column due to gain guiding and refraction anti-guiding.

49.3.9 Peak Spectral Brightness

Soft X-ray lasers are among the brightest soft X-ray sources available. Both laser-excited [64] and discharge-pumped collisional excitation lasers have reached brightness values that exceed $1 \times 10^{25} \text{ photons mm}^{-2} \text{ mrad}^{-2} \text{s}^{-1} (0.01\% \text{BW})^{-1}$ [79]. Figure 49.2 compares the peak spectral brightness of some of the brightest soft X-ray lasers with other sources of coherent short-wavelength radiation.

49.3.10 Average Spectral Brightness

Due to their low-repetition rate, most of presently available saturated soft X-ray lasers produce low-average brightness. An exception is a capillary discharge 46.9 nm laser [79] that produced an estimated spatially coherent average output power of nearly 1 mW within a relative spectral bandwidth ($\Delta \lambda/\lambda < 1 \times 10^{-4}$), a spectral purity that exceeds that of some undulators at third-generation synchrotrons. The average spectral brightness of laser-pumped soft X-ray lasers can be expected to increase significantly in the near future as compact high-repetition rate optical laser pumps become available and more efficient target configurations are used to couple the laser pump energy to the plasma.

49.4 Other Methods for the Generation of Coherent X-Ray Radiation

The direct amplification of radiation in plasmas is not the only means by which coherent soft X-ray radiation can be generated. Alternative methods include high-order harmonic generation from the output of high-power optical lasers [80–84], synchrotron sources [85–88], free-electron lasers (FELs) and up-conversion of optical laser pulses by scattering with energetic electron beams [89–92]. Synchrotron sources are large multi-user facilities based on the radiation emitted by high-energy electron beams. They have the advantages of broad tunability and high-average power. However, they fall
short of the high peak brightness needed in applications such as the study of non-linear phenomena at ultrashort wavelengths [93,94] and the diagnostics of dense plasmas [29,30,95,96]. Another alternative, also based on accelerator technology, is the use of self-amplified spontaneous emission in an FEL [88,89]. In a soft X-ray FEL, an electron beam would radiate at much higher powers and with better coherence than it does due to spontaneous synchrotron radiation. Several single-pass ultrashort-wavelength FELs have been proposed or are under construction [88,90]. Nevertheless, these lasers will also constitute large facilities that will not be tabletop ones.

Alternatively, soft X-ray coherent radiation can also be generated using non-linear optical techniques for frequency up-conversion of optical laser radiation. Harmonic up-conversion of intense ultrashort pulse tabletop optical laser systems has been reported to generate radiation at wavelengths as short as 2.7 nm [80]. Presently, fairly optimized conditions in non-phase-matched configurations typically yield a conversion efficiency of about 10⁻⁶ in the range of 10⁻⁹ to 40 eV (of the order of 10⁸ photons per pulse) and 10⁻⁸ in the 40–150 eV range (10⁶–10⁷ photons per pulse) [81]. The highest energy reported for a high-order harmonic pulse, 60 nJ at about 50 eV, was obtained using a powerful glass laser [82]. The demonstration of phase-matched harmonic conversion of visible light into soft X-rays with a generation efficiency of 10⁻⁵–10⁻⁶ in the 40–70 eV spectral region was reported [83,84]. Soft X-ray pulses with an energy >0.2 nJ pulse⁻¹ per harmonic order were produced at a repetition frequency of 1 kHz utilising 20 fs optical pulses [83].

49.5 X-ray Laser Schemes

49.5.1 Collisional Electron Excitation Lasers

49.5.1.1 Quasi-steady-state Lasers

Collisional electron impact excitation of Ne-like and Ni-like ions has resulted in some of the most robust soft X-ray lasers available. In the traditional implementation of these lasers, the generation of a population inversion occurs in a quasi-cw regime by strong collisional monopole electron excitation of the upper level aided by the very favourable ratio between the radiative lifetime of the upper and lower levels. The upper levels are metastable with respect to radiative decay to the ground state, and the lower levels are depopulated by strong dipole-allowed transitions. The first successful demonstration of a collisional soft X-ray laser was realized at Lawrence Livermore National Laboratory and involved the 2p^53p → 2p^53s transitions in Ne-like Se and Ne-like Y [17]. Lasing has been extended to nearly all of the Ne-like ions with an atomic number between Si [97] and Ag [98] at wavelengths ranging from 87 to 9.93 nm.

Figure 49.3 shows a simplified energy-level diagram for a typical Ne-like system (GeXXIII) illustrating the laser transitions and the dominant processes involved in the generation of amplification. The 3p upper levels are dominantly populated by electron monopole collisional excitation (E_c) from the Ne-like ion ground state and by cascades from higher energy levels. The population inversions are maintained by the very rapid

![Figure 49.3](From Keane [50].)
radiative decay of the 3s lower levels to the ground state of the ion through strong dipole-allowed transitions ($E_1$). Therefore, operation of these lasers in a quasi-cw regime requires the plasma to be optically thin for the transitions originating from the lower level.

Lasers in the $3d^{9}4d \rightarrow 3d^{9}4p$ transitions of Ni-like ions are direct analogues to lasers in $2p^53p \rightarrow 2p^53s$ transitions in closed-shell Ne-like ions but have the advantage of producing amplification at shorter wavelengths for a given state of ionization. This higher quantum efficiency significantly reduces the pumping energy required to achieve lasing of a selected wavelength. Ni-like soft X-ray lasers were first demonstrated in 1987 in an Eu-laser-created plasma, producing amplification with a $g \ell \approx 4$ at 7.1 nm [99]. Subsequently, the scheme was isoelectronically extrapolated to other ions with laser wavelengths as short as 3.56 nm in Ni-like Au [100]. Gain-saturated operation has been obtained at wavelengths as short as 5.8 nm [21]. Hagelstein first proposed the use of low-Z Ni-like ions to develop tabletop collisional lasers at wavelengths near 20 nm and computed a significant gain for overheated plasma conditions [101]. Gain has also been observed in Co-like ions [100], and the use of the Nd-like sequence has also been proposed [101].

The collisionally excited soft X-ray lasers developed before 1990 consisted of line focus plasmas generated by optical laser pulses with energies ranging from several hundred joules to several kilojoules and, therefore, involved the use of very large facilities [17,20,97–100,102–106]. One such laser is schematically illustrated in Figure 49.4. Much progress has been made in improving laser efficiency. Strong refraction of the amplified beam caused by the large electron density gradients in the amplifier was early recognized as a major obstacle to the generation of efficient soft X-ray lasers with good beam quality. Methods implemented to mitigate refraction have included the use of foil targets [48,107–110], opposite-gradient and curved targets and, in particular, the use of one or multiple pre-pulses [111–130]. In the pre-pulse technique, the first pulse is used to create a plasma with an optimized density for amplification and with reduced density gradients for improved beam propagation. The subsequent pulses which are absorbed more efficiently in the gain region to heat the plasma to lasing conditions lead to the observation of large amplification in a large number of elements with dramatic increase in the $(J=0) \rightarrow (J=1)$ line intensity and reduced excitation energy.

Another important step in the reduction of the pump energy has been the use of shorter excitation pulses. Daido et al. [131,132] reported lasing in several lanthanide ions in the spectral range 5.8–14 nm using a three-pre-pulse sequence and a main excitation pulse of 100 ps and ~250 J of energy. A subsequent series of experiments conducted at the Rutherford Laboratory obtained saturation in several transitions [129,133–135] with wavelengths as short as 5.86 nm (Ni-like Dy [136]) using sequences of 75 ps pulses with about 100J of energy saturation. Balmer et al. have used a relatively compact Nd:glass laser to demonstrate saturated lasing in Ne-like Fe (25.5 nm), Ni-like Ag (14.0 nm) and Ni-like Pd (14.7 nm) with driver energies below 30 J in a 100 ps pulse [137,138]. In terms of improving the soft X-ray output beam characteristics, double-pass amplification experiments have demonstrated increased spatial coherence and reduced beam divergence.
### 49.5.1.2 Transient Collisional Lasers

The collisional electron excitation scheme described earlier is intrinsically a quasi-steady-state scheme in which lasing can occur for as long as the plasma conditions necessary for the generation of a population inversion can be maintained. It was first recognized by Afanasiev and Shlyaptsev [139] that one to two orders of magnitude larger gain coefficients could be produced for a short period of time (typically subpicosecond to tens of picoseconds) by heating the plasma at a rate faster than the relaxation rate of the excited states. The larger gain coefficients are mainly the consequence of the larger rate of excitation of the upper level from the ion ground state by electron collisions that result in a large population inversion before collisions have time to redistribute the populations. Another phenomenon that contributes to an increased gain is the increased rate of electron excitation in an overheated plasma. Transient gains in excess of 100 cm\(^{-1}\) have been predicted theoretically [139, 140]. In the transient regime, there is no need to limit the transverse dimension of the plasma in order to ensure optical transparency of the laser lower level radiation. A main advantage of the transient excitation scheme for the realization of tabletop X-ray lasers is the greatly reduced pump energy required for excitation.

The availability of multi-terawatt ultrashort pulsed optical laser systems with output energies of several joules has provided the opportunity to demonstrate soft X-ray lasing by transient electron collisional excitation [60–66]. The implementation is based on a two-step excitation sequence. First, a long pulse (typically of nanosecond duration) is used to produce a plasma containing the desired active ions, which are usually closed-shell Ne-like or Ni-like. The temperature of this pre-plasma must be sufficiently high to produce a ground-state population of these ions but it does not need to reach the values necessary to populate the upper level. The plasma is allowed to expand hydrodynamically to reach the desired degree of ionization, optimum electron density and minimum possible electron density gradient. Second, the plasma is rapidly heated with a picosecond or subpicosecond laser pulse to increase the electron temperature rapidly to values that exceed the excitation energy of the upper level. The ionization balance is not significantly altered and a large transient population inversion is generated by electron excitation.

The first demonstration of amplification by transient inversion was realized by Nickles et al. in the 32.6 nm line of Ne-like Ti [60]. The experiment used a hybrid chirped-pulse amplification (CPA) Ti:sapphire/Nd:glass pump delivering synchronized long and short pulses of ~1.2 ns and 0.7 ps duration and energy of 3 and 2 J, respectively. The soft X-ray pulse duration was measured to be less than 20 ps. The results were analysed to correspond to an average gain of 19 cm\(^{-1}\) and to \(g_I \approx 9.5\) [60]. Weaker lasing was also observed in a line near 30 nm, identified as the \(3d(J=1) \rightarrow 3p(J=1)\) transition of Ne-like Ti. These results were improved and extended to other Ne-like ions, such as Fe and Ge, in subsequent experiments conducted at several laboratories [61–66].

The transient collisional scheme has also been extended to shorter wavelengths utilizing the Ni-like sequence [61–66]. Dunn et al. reported a gain of up to 35 cm\(^{-1}\) and \(g_I \approx 12.5\) in the \(4d^5S_0 \rightarrow 4p^1P_1\) line of Ni-like Pd at 14.7 nm and large amplification in several other Ni-like ions [61] using the relatively compact laser system illustrated in Figure 49.5. The pump laser occupies two 1.2×3.6 m optical tables with a total area of less than 10 m\(^2\). This 15 TW hybrid laser system consists

---

**FIGURE 49.5** Schematic illustration of the CPM pump laser system and target chamber used to produce transient inversion collisional soft X-ray lasers at Lawrence Livermore National Laboratory. The pump laser occupies two standard optical tables of dimensions 1.2×3.6 m with total area less than 10 m\(^2\). The system can be fired once every 4 min. (Courtesy of J Dunn, Lawrence Livermore National Laboratory.)
of a Ti:sapphire oscillator tuned to 1050 nm, and four-stage Nd-phosphate glass amplifiers. It generates two synchronized laser pulses of 7.5 J in 500 fs and 15 J in 600 ps at a repetition rate of one shot every 4 min. Typically 50 shots on target are achieved in a day. The full circles in Figure 49.6b show the measured increase of the Ni-like Pd 14.7 nm line intensity for target lengths between 2 and 9 mm. The gain coefficient for lengths between 1 and 2 mm is about 35 cm\(^{-1}\) but continuously decreases to reach a value of 3.9 cm\(^{-1}\) for target lengths above 7 mm. This smooth decrease in gain with target length has been observed in all non-travelling-wave transient collisional excitation experiments and resembles gain saturation. However, it is mainly caused by the short duration of the gain and by refraction. Gain saturation with the transient excitation scheme was first demonstrated on the Ne-like scheme for the 32.6 nm line of Ne-like Ti and the 19.6 nm line of Ne-like Ge at the Rutherford Laboratory [63]. However, these experiments utilized a total reported excitation energy of 32 and 60 J, respectively, which is not available in smaller facilities. To achieve gain saturation with smaller excitation energy, travelling-wave excitation schemes have been implemented at several laboratories.

### 49.5.1.3 Travelling-wave Excitation

In these transient systems, the shortness of \(\tau_g\), the gain duration, limits the amplification length to \(l < \frac{c \tau_g}{\omega}\) (where \(c\) is the speed of light in the plasma), unless travelling-wave excitation is used to maintain the excitation in phase with the amplified X-ray pulse. The travelling-wave system was used to demonstrate gain saturation in Ni-like Pd at 14.7 nm with 1.8 J of long pulse energy and 5.2 J of short pulse. Figure 49.7 illustrates the advantage of travelling-wave excitation and corresponds to a 20–100-fold increase in X-ray laser intensity compared to the configuration without travelling-wave excitation [65]. The soft X-ray laser output energy with travelling-wave excitation was estimated at \(\sim 10\ \mu J\). Strong amplification was also obtained in Ni-like ions ranging from Mo (18.9 nm) to Sn (11.9 nm) [63]. A different travelling-wave method was also used at the Rutherford Laboratory to obtain gain saturation in several Ne-like ions and in Ni-like Sm at 7.3 nm [65]. Experiments conducted at CEA-Limeil yielded saturation of the 13.9 nm laser line of Ni-like Ag and also observed strong amplification at 16.05 nm in a 4f–4d transition in the same ion [66].

Table 49.1 compares the characteristics of soft X-ray lasers driven by the transient collisional scheme at a facility with a relatively compact multi-terawatt laser with those of the quasi-steady-state collisional lasers pumped by the Nova laser facility [64]. Future improvements in pumping efficiency resulting from optimized target configurations and travelling-wave excitation can be expected to lead to saturated transient inversion

![Figure 49.6](image1.png)

**FIGURE 49.6** Results of transient inversion soft X-ray amplification experiment in the 4d–4p \(J=0\rightarrow 1\) line of Ni-like Pd at 14.7 nm. (a) Spectra showing the increase of the intensity of the laser line as a function of target length. The intensity increases by more than three orders of magnitude when the target length is increased from 1 to 3 mm. (b) Measured intensity of the laser line as a function of target length. The continuous line is a guide through the experimental points. The data show the decrease of the transient gain as the X-ray laser propagates along the plasma column. (c) Computed transient gain profiles as a function of time measured from the arrival of a 1 ps excitation pulse. (From Dunn et al. [61].)

![Figure 49.7](image2.png)

**FIGURE 49.7** Comparison of the X-ray laser intensity versus plasma column length for the Ni-like Pd 4d–4p laser line at 14.7 nm with travelling wave (closed circles) and without travelling wave (open circles). The excitation energy consisted of a 1.8 J long pulse and a 5.2 J short pulse. (From Dunn et al. (2000) [64].)
TABLE 49.1
Characteristics Transient Inversion Collisional Laser Pumped by a Compact Multi-terawatt Laser, as Compared with Quasi-Steady-State Collisional Laser Pumped by NOVA (after Dunn et al. [141])

<table>
<thead>
<tr>
<th></th>
<th>COMET</th>
<th>NOVA</th>
</tr>
</thead>
<tbody>
<tr>
<td>Size (sq. ft)</td>
<td>100</td>
<td>&gt;40 000</td>
</tr>
<tr>
<td>Cost ($)</td>
<td>1 M</td>
<td>&gt;100 M</td>
</tr>
<tr>
<td>Pump energy (J)</td>
<td>&lt;10</td>
<td>5–10 × 10^3 (2 beams, 1α)</td>
</tr>
<tr>
<td>XRL gain (cm⁻¹)</td>
<td>30–65</td>
<td>1–8</td>
</tr>
<tr>
<td>XRL output (J)</td>
<td>&gt;10 × 10⁻⁴ (at 146.8 Å)</td>
<td>5 × 10⁻³ (at 155 Å)</td>
</tr>
<tr>
<td>Shot rate (per day)</td>
<td>50–100</td>
<td>4–6</td>
</tr>
<tr>
<td>XRL wavelength (Å)</td>
<td>119–330</td>
<td>35–330</td>
</tr>
<tr>
<td>Pulse duration (ps)</td>
<td>5–10</td>
<td>45–200</td>
</tr>
<tr>
<td>Brightness (ph mm⁻² mrad⁻² s⁻¹ (0.01% BW)⁻¹)</td>
<td>10⁻¹⁴</td>
<td>10⁻¹⁴</td>
</tr>
<tr>
<td>Cost/shot ($)</td>
<td>50–100</td>
<td>10 000–20 000</td>
</tr>
</tbody>
</table>

soft X-ray lasers occupying a single optical table and to tabletop lasers that operate at shorter wavelengths.

Another method for pumping collisional lasers in which travelling-wave excitation is intrinsic was demonstrated by Lemooff et al. in Pd-like Xe utilising optical field ionization. In this scheme, an intense circularly polarized femtosecond laser pulse is used to simultaneously create closed-shell ions and hot pumping electrons by tunnelling ionization [59]. An amplification of gl ≈ 11–12 in the 41.8 nm line of Pd-like Xe was measured by axially pumping a Xe gas cell up to 8.4 mm in length with 70 mJ pulses from a Ti:sapphire laser operating at 10 Hz.

49.5.1.4 Collisionally Excited Capillary Discharge Lasers

Direct excitation of plasma columns with an electrical discharge has the advantage of generating soft X-ray lasers that are very efficient and compact. Fast-discharge excitation of capillary plasmas has produced the highest soft X-ray laser average power to date [44,45]. In these lasers, the electromagnetic forces of a fast rising current pulse flowing through a capillary channel rapidly compresses the plasma to a size of about 300 µm diameter. The fast current rise time minimizes the amount of material that is ablated from the capillary walls before the magnetic field detaches it from the walls [36–39]. The first observation of large soft X-ray amplification in a discharge-created plasma was realized in 1994 by Rocca et al. in the 46.9 nm line of Ne-like Ar [37,38]. In that initial experiment, illustrated in Figure 49.8, the fast capillary discharge set-up (generating current pulses of 60 ns half-cycle duration and ~40 kA peak current) was used to excite Ar plasma columns in 4 mm diameter capillary channels up to 12 cm in length. As indicated in Figure 49.9, the capillary was placed in the axis of a 3 nF liquid-dielectric capacitor that was pulse-charged by a Marx generator. The capillary loads were excited by discharging the capacitor through a spark-gap switch pressurized with SF₆. A small gain was also observed in the (J=2) → (J=1) line of Ne-like Ar at 69.8 nm [38,136]. The optimum conditions for lasing occur several nanoseconds before stagnation, when the first compression shock wave reaches the axis. Lasing by collisional electron excitation of Ne-like Ar ions takes place at a time when the electron density is rapidly increasing and reaches (0.3–1 × 10¹⁰ cm⁻³), when the electron temperature is 60–80 eV [40]. The pulsewidth is approximately 1 ns [40,43]. Subsequent experiments employing longer plasma columns under better optimized discharge conditions yielded an effective gain–length product of 27 and resulted in the first observation of gain saturation in a tabletop soft X-ray amplifier [40]. Saturation of the laser intensity was observed at products of about 14 [40].

A very compact, high-repetition rate, saturated 46.9 nm laser of a size comparable to that of many widely utilized visible and ultraviolet gas lasers has been developed [44,45]. Figure 49.10 illustrates the size of this capillary discharge soft X-ray laser in comparison to a 1 J Nd:YAG laser. The soft X-ray laser occupies a surface space of about 1 m by 0.4 m on an optical table. This laser was operated with capillaries 18.2 cm in length at a repetition rate of 7 Hz to produce an average output pulse energy of 135 µJ, corresponding to an average laser power of 1 mW [44]. Increasing the plasma column length to 35.4 cm resulted in the generation of 0.88 mJ laser pulses at a repetition rate of 4 Hz [45], corresponding to an average power of 3.5 mW as shown in Figure 49.11. The spatially coherent average output power per unit bandwidth emitted by this compact laser at 26.5 eV is comparable to that generated by a beam-line at a third-generation synchrotron facility, while its peak coherent power per unit bandwidth exceeds that of the synchrotron by several orders of magnitude [44,45]. Table 49.2 summarizes the characteristics of the 46.9 nm Ne-like capillary discharge laser. These capillary-discharge-pumped tabletop lasers were successfully used in several applications, including high-resolution soft X-ray laser interferometry and shadowgraphy of plasmas [95,96,142,143], the measurements of XUV optical constants of materials [144] and the demonstration of laser ablation with a focused soft X-ray beam [145].

The Ne-like Ar results were extended to Ne-like S (60.8 nm) [42] and Ne-like Cl (52.9 nm). To obtain amplification in Ne-like S, the discharge set-up illustrated in Figures 49.8 and 49.9 was modified to allow for the injection of the sulphur vapour into the capillary channel through a hole in the ground electrode. The sulphur vapour was produced, ablating the wall of an auxiliary capillary channel, drilled in a sulphur rod, with a slow current pulse. Model computations indicate that, in this
In this population inversion scheme, first suggested by Gudzenko and Shelepin [147], the upper level is populated following the recombination of ions of atomic species A with a charge \(Z+1\) with an electron, through a three-body interaction described as collisional or three-body recombination:

49.5.2 Collisional Recombination Lasers

49.5.2.1 Mechanisms

In this population inversion scheme, first suggested by Gudzenko and Shelepin [147], the upper level is populated following the recombination of ions of atomic species A with a charge \(Z+1\) with an electron, through a three-body interaction described as collisional or three-body recombination:
This process preferentially populates highly excited bound levels \((A^{Z+1})\) of the ion of charge \(Z\), favouring the generation of population inversion. The collisional recombination rate is proportional to the square of the electron density. The recombination rate is also extremely sensitive to \(T_e\), the electron temperature, following a dependence on \(T_e^{-4.5}\). Therefore, the generation of large population inversions by recombination requires a dense and relatively cold plasma.

Hydrogen-like ions have a very favourable energy-level structure for the generation of population inversions by collisional recombination. In principle, several transitions can be inverted but, initially, much of the attention focused in the very favourable \(3 \rightarrow 2\) transition. Figure 49.12 shows the atomic processes involved in the generation of population inversion in the \(3 \rightarrow 2\) transition of H-like C at 18.2 nm. When the plasma cools, collisional recombination populates highly excited states and electron collisions rapidly transfer the population to levels of lower energy. Since collisional electron de-excitation is inversely proportional to the square root of the energy difference between the levels, this electronic cascade reaches a level at which electron de-excitation is no longer dominant over radiative decay. At this level, the bottleneck that is produced in the cascade creates a population inversion with respect to a lower level that is de-excited by very rapid radiative decay to the ion ground state.

The amplification of soft X-rays by plasma recombination would seem to require conflicting plasma conditions: a very highly ionized plasma and a very cold electron temperature. In practice, the problem has been traditionally solved utilizing a two-step process. First, a hot and dense plasma with a large population of \(A^{Z+1}\) ions is generated by a heating pulse. Second, the plasma is rapidly cooled by an adiabatic expansion [10,11,16,148–155], by electron heat conduction to a nearby wall or colder neighbouring plasma [156] or by radiation from high-Z ions introduced as impurities into the plasma [157,158]. All three cooling mechanisms, or combinations of them, have been utilized experimentally to generate gain at soft X-ray wavelengths by collisional electron–ion recombination.

Important efforts have been devoted to amplification in the 18.2 nm \(3 \rightarrow 2\) line of H-like C. Initial experiments observed population inversions in plasmas generated by ablating solid carbon targets or carbon fibres with high-power laser pulses and cooled by adiabatic expansion [10,11,14–16]. Large amplification was first observed by Suckewer et al. in an experiment in which a 300 J pulse from a CO\(_2\) laser with about 75 ns pulse-width was used to generate a nearly totally ionized carbon plasma column by bombarding a carbon solid target immersed in a strong solenoidal magnetic field [18]. The magnetic confinement allowed the maintenance of a high electron density while the plasma was cooled by radiation and electron heat conduction to adjacent cooling blades. Laser pulses of about 3 mJ were generated at an efficiency of \(10^{-3}\). Amplification has also been demonstrated in numerous experiments in which line focus plasmas were cooled by free adiabatic expansion [30,149–154]. Gain has been reported in H-like Na ions [152], in the \(4f \rightarrow 3d\) and \(5f \rightarrow 3d\) lines of Li-like ions [153,154] and in Be-like and Na-like ions [159].

Several experiments have been mostly conducted toward the realization of a small-scale recombination laser [160–167]. In 1989, Kim et al. reported a gain of about 8 cm\(^{-1}\) in the 18.2 nm line of CVI using a 25 J pump pulse of 3 ns duration, line-focused into a 5 mm-long solid carbon target [160]. Later experiments obtained similar gain coefficients target with only 4 J of laser excitation energy using a multi-fin carbon target [162]. However, difficulties were found in increasing the amplification over \(gl \approx 5\). One of the latest attempts to increase the product in this line utilized a polyethylene microcapillary as target [163]. A Nd:YAG laser (2.5 J, 1.5 ns pulsewidth) was focused at the entrance of the microcapillary with the motivation of guiding the pump beam, plasma confinement and plasma cooling by heat conduction to the walls. A product of

\[
(A^{Z+1}) + 2e \rightarrow (A^Z)^+ + e. \tag{49.10}
\]
about 5 was inferred from the intensity ratio of lines in the axial and perpendicular direction. A similar experiment conducted in B$_2$O$_3$ resulted in a gain of $g_l \approx 5$ in the $3 \rightarrow 2$ transition of H-like B at 26.2 nm [167]. A pre-plasma was created by a 20 ns, 0.2 J KrF laser; and the excitation was produced by a 0.4 J, 8 ns Nd:YAG laser operating at 1 Hz. Hara et al. have utilized multiple-pulse excitation from a pulse train with the objective of developing a compact soft X-ray recombination laser based on a train of 100 ps pulses produced by a tabletop Nd:YAG laser [164–166]. Experiments conducted utilising 1.5–2 J of laser pulse energy to excite an 11 mm long Al slab target were reported to produce a gain coefficient of 3.2 cm$^{-1}$ in the 15.5 nm line of Li-like Al [166] but the amplification was small.

An advantage of the recombination scheme with respect to the collisional excitation scheme involving $\Delta n = 0$ transitions is its more rapid scaling to shorter wavelengths with nuclear charge $Z$. However, recombination lasers have suffered the problem of not scaling adequately with plasma column length, a problem possibly related to the very high sensitivity of the gain in recombination schemes to the variation of the plasma parameters. The largest product achieved to date is 8 [18].

### 49.5.2.2 Gain in Discharge-pumped Recombination Systems

The possibility of obtaining amplification by collisional recombination in capillary discharge plasmas was proposed by Rocca et al. in 1988 [168]. Evidence of gain has been reported in several discharge-pumped recombination laser experiments [169–175]. However, in all cases, only small exponentiation ($g_l < 4$) has been observed to date, with the line intensity remaining of the same order as that of surrounding non-lasing lines emitted by the plasma.

![FIGURE 49.11 Measured output pulse energy and average output power of a tabletop capillary discharge 46.9 nm laser operating at a repetition frequency of 4 Hz. (a) Shot-to-shot laser output pulse energy. (b) Average output power computed as a walking average of 60 contiguous laser pulses. (c) Distribution of the output pulse energy. The average pulse energy is 0.88 mJ, corresponding to an average power of 3.5 mW. (From Macchietto et al. [46].)](image)

![FIGURE 49.12 Simplified Grotrian diagram of H-like C showing the processes responsible for the generation of population inversion between the $n = 3$ and $n = 2$ levels in the 18.2 nm CVI recombination laser.](image)

| Characteristics of Capillary-discharge-pumped Tabletop 46.9 nm Laser in Ne-Like Ar |
|---------------------------------|-----------------|
| **Laser Parameters**           | **Ref.**        |
| Pulse energy                   | 0.88 mJ at 4 Hz | [46] |
| Average pulse power            | 3.5 mW          | [46] |
| Peak pulse power               | 0.6 MW          | [46] |
| Divergence                     | ≈4.6 mrad       | [45,46] |
| Pulswidth                      | 1.2–1.5 ns      | [45,46] |
| Peak spectral brightness       | $2 \times 10^{21}$ photons s$^{-1}$m$^{-2}$ | [79] |
|                                | mrad$^{-2}$ (0.01% bandwidth)$^{-1}$ |
49.5.2.3 Recombination Lasing in Transitions to the Ground State

Jones and Ali [9] demonstrated theoretically that large transient gains could be generated in the $2 \rightarrow 1$ transitions of H-like ions following recombination of a totally ionized plasma of arbitrarily low temperature. This kind of laser in a transition to the ground state is very attractive to rapidly scale recombination lasers to very short wavelengths. However, for lasing to occur in a transition to the ground state, recombination has to be very rapid to allow for the generation of large upper level populations before the ion ground-state level is significantly populated. The recombination time must then be shorter than the radiative lifetime of the upper level which is, for example, 26 and 1.6 ps for the $n=2$ level of H-like Li and H-like C, respectively. These time scales are shorter than the time required to cool a hot plasma before the ground state is significantly populated. Burnett and Corkum [176] recognized that a cold plasma of highly stripped ions in which recombination can occur in a very short time scale can be created by optical-field-induced ionization (OFI) using linearly polarized light and proposed the realization of ultrashort wavelength lasers utilizing this approach. Several theoretical studies analysed different aspects of the implementation of soft X-ray recombination lasers based on OFI [177,178].

The first report of amplification following plasma recombination of an OFI plasma corresponds to an experiment 13.5 nm in the $2 \rightarrow 1$ transition of H-like Li realized by Nagata et al. in 1993 utilizing the set-up illustrated in Figure 49.13a. [57]. A cold plasma of totally ionized Li ions was generated in two steps. First, a plasma of singly-ionized Li ions was produced by line-focusing a 20 ns KrF laser pulse with an energy of 200 mJ onto a rotating lithium target. Second, after a selected time delay, the plasma was optical-field ionized with 50 mJ pulses from a linearly polarized subpicosecond (0.5 ps) KrF laser pulse focused to an intensity of $1 \times 10^{17}$ cm$^{-2}$. Figure 49.13b shows the measured variation of the 13.5 nm laser line intensity with plasma length, from which a gain coefficient of 20 cm$^{-1}$ was deduced. The 13.5 nm radiation pulse was measured to have a FWHM duration of less than 20 ps, with the peak intensity occurring about 20 ns after the pump pulse. However, the maximum resulting product only reached four because the plasma length was limited to 2 mm by ionization-induced refraction. This limitation is the result of the confocal geometry that causes the beam to refract due to the convex electron density profile resulting from the maximum ionization on axis. Approaches to overcome this problem have been suggested and include the generation of very flat transverse intensity profiles [178] and the generation of plasma waveguides with a minimum density on axis [58,179–184].

Korobkin et al. [58] used the set-up illustrated in Figure 49.14a to increase the amplification length to 5 mm by pre-forming a plasma waveguide in an LiF microcapillary tube. A plasma with a minimum electron density on axis was generated by ablation of the capillary walls with a 100 mJ, 5 ns pulse of 1.06 µm wavelength radiation from a Nd:YAG laser loosely focused on the entrance of the microcapillary. Following a time delay of several hundred nanoseconds, the plasma was further ionized with a laser pulse of 50 mJ energy and 250 fs duration from a tightly focused KrF laser. The observed increase of the intensity of the 13.5 nm line of H-like Li as a function of length is shown in Figure 49.14b, and corresponds to $g_l=5.5$ [57]. These results were obtained at a repetition rate of 2 Hz. Gain by recombination in an OFI plasma has also been reported in the 2p2s3P$\rightarrow$2p2P transition of OIII ($\lambda=37.4$ nm) and in the 2p3s3P$\rightarrow$2p5D line of OII ($\lambda=61.7$ nm) [185]. The data suggest gain coefficients as high as 25 cm$^{-1}$ but again the overall amplification was small due to the small length of the gas targets.

The small excitation energies used in OFI soft X-ray laser experiments are compatible with the requirement for the generation of high-repetition-rate tabletop soft X-ray lasers. However, an increase in the product from the reported values of ~6.5 to the larger values is still required to achieve practical output energies. Predictions of the output pulse energies that can be expected from OFI lasers range from 1 µJ [178] to 10–20 µJ [186]. The OFI recombination scheme can, in principle, also be scaled to shorter wavelengths by using ions with higher charge and more intense optical fields, provided that parametric electron heating effects (mainly stimulated Raman backscattering) can be overcome.

FIGURE 49.13 (a) Schematic illustration of the experimental set-up used at RIKEN in the observation of amplification in the 13.5 nm line of H-like Li in a plasma created by optical field induced ionization. (b) Results of amplification experiment in the $2 \rightarrow 1$ line of H-like Li in a plasma created by optical-field-induced ionization. Intensities of the 13.5 nm laser line of H-like Li and 19.9 nm line of He-like Li as a function of plasma column length. The broken curve is a calculated increase corresponding to a small-signal gain coefficient of 20 cm$^{-1}$. (From Nagata et al. [57]).
49.5.3 Photoionization Lasers

The generation of amplification following the selective X-ray photoionization of inner shell electrons was originally proposed by Duguay and Rentzepis in 1967 [1]. The generation of population inversions by this mechanism exploits the fact that at photon energies just above the threshold for inner-shell photoionization, the cross section is an order of magnitude larger for inner-shell electrons as compared to outer-shell electrons. This scheme has the potential advantage of leading to compact lasers with wavelengths shorter than 1.5 nm [187,188].

In principle, it can allow for operation at a low plasma temperature (<1 eV) with consequently less Doppler broadening and large gain coefficients. The incoherent X-ray photons that pump the laser media can be produced by a nearby plasma created by heating a high-Z target material, such as gold, with an intense ultrashort laser pulse. Photons at energy below the inner-shell binding energy can be removed from the pump with an appropriate filter to avoid pumping of the lower level.

The first proposal for inner-shell photoionization lasers at X-ray wavelengths [2] preceded by decades the development of sufficiently powerful ultrashort pulse laser drivers. McGuire proposed the production of population inversions by selective Auger decay [189]. In 1986 Kapteyn et al. demonstrated lasing at 108.9 nm in doubly ionized Xe by Auger decay following the photoionization of neutral Xe [190]. Output energies of several µJ in this transition were subsequently obtained at a 2 Hz repetition rate by Sher et al. utilizing travelling-wave excitation [191]. Amplification in the equivalent transition in Kr at 90.7 nm was also observed [192]. However, no demonstration of a photoionization laser at X-ray wavelengths has yet been realized. Nevertheless, the development of lasers capable of producing peak powers of up to 100 TW with ~20 fs pulse duration increases the likelihood that this type of X-ray lasers will soon be realized.

Kapteyn has analysed the possibility of obtaining lasing by preferentially photoionizing the K-shell electrons of low-Z elements [187], focusing in particular on the Kα line of Ne (λ=1.5 nm). A main advantage of the Kα scheme is its scalability to very short wavelengths. The energy-level diagram of the Kα Ne laser is illustrated in Figure 49.15. The filtered X-rays from the pump primarily photoionize the inner-shell electrons, producing population inversion and gain in the allowed radiative transition between the (1s)−12S and (2p)−12P levels of the singly charged ion. Rapid Auger decay with a rate of (2.7 fs)−1 is the dominant mechanism of depopulation of the (1s)−1 upper level. However, the self-terminating lifetime is not the dominant process that limits the duration of the gain.
Instead, the magnitude and duration of the gain is limited by electron collisional ionization of neutral neon atoms. The energetic electrons created in the lasing material by photoionization and Auger decay create predominantly ground-state Ne ions that are the lower level, destroying the gain. Therefore, this is intrinsically an ultrashort pulse X-ray laser. It has been estimated that lasing in the $K\alpha$ transition of Ne at 1.5 nm could be produced by travelling-wave excitation using a pump laser generating $\approx 10^9$ pulses of approximately 50 fs duration [187]. Eder et al. [178,188,193] calculated a scaling law relating the necessary laser pump energy $E_\text{L}$ (J) to the wavelength $\lambda$ (nm) that can be achieved. Assuming 20 fs X-ray pump pulses, they obtained $E_\text{L} = (4.5/\lambda)^2$ [194]. Calculations for lasing in C at 4.5 nm suggest that driving laser energy of the order of 1 J should be sufficient to produce a gain coefficient of 10 cm$^{-1}$ and a large product [193].

The possibility of using inner-shell electrons to obtain lasing in $2p \rightarrow 2s$ Ne-like ions and $3d \rightarrow 3p$ Ni-like ion transitions has also been suggested [195,196]. Barty et al. and Kim et al. have discussed systems in which dominant Coster–Kronig decay of the laser lower level can overcome electron collisional filling of this state [197–204]. These systems are, therefore, more immune to secondary electron ionization. Computations show that, under certain conditions, inversions can be obtained with pumping by energetic electrons as well as X-rays. Conditions for inversion by Coster–Kronig lower level decay were analysed in several transitions in elements with nuclear charge up to $Z=90$, and detailed computations of gain were published for the $L_2-M_1$ transition in Ti ($Z=22$) at 3 nm.

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