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X-RAY LASERS : STATE OF ART AND PROSPECT

P. JAEGLE

Laboratoire de Spectroscopic Atomique et Ionique, Universite Paris-Sud, F-91405 Orsay Cedex, France
and Greco "Interaction Laser-Matiere", Ecole Polytechnique, F-91128 Palaiseau Cedex, France

Abstract

Using hot plasmas for making soft X-ray amplifiers will lead to laboratory sources brighter than any others by several orders of magnitude. Population inversions are produced either by collisional pumping due to plasma free electrons or by fast plasma cooling. Gain coefficients between 1 and 5 per cm have been measured in the 60 A - 200 A wavelength range.

Introduction

During the last years, one has realized the capability of hot plasmas to act as amplifiers in the X-ray range /1,2/. A familiar idea is the basic requirement of any laser to be the existence of population inversion between two atomic or molecular states, in a gaseous or solid material, so that decay by stimulated emission may yield amplification. The production of population inversion needs pumping energy. In the case of X-rays, owing to the short life-time of excited levels, the difficulty was that a large amount of pumping energy must be brought into the active medium in a very short time. Therefore a huge power was required if the usual pumping techniques were to be used. Now the fact is that hot plasmas can give rise to population inversion and stimulated emission just in consequence of a suitable matching of atomic physics and hydrodynamics, i.e. without external pumping. This has kept out the great difficulties associated with the optical pumping of an X-ray laser. We will explain the main physical mechanisms which make that possible and lead to two major types of inversion schemes, i.e. the recombination in cooling plasma and the free electron collisional pumping. Then we will shortly describe the principle of soft X-ray laser experiments and the results obtained so far. We will conclude in comparing X-ray lasers with other sources regarding the brightness and possible applications.

Plasmas for soft X-ray lasers

In hot plasmas the electronic temperature is comprised between 100 ev and several keV (1 ev corresponds to 11605*K). The X-ray emission comes from optical transitions between the states of multicharged ions. Z being the charge of the ion core, i.e. the number of removed electrons, and n the main quantum number, in the simplest case the energy of the level n is related to Z by:

\[ E_n = -13.6 \times Z^2/n^2 \text{ ev} \]

In addition to discrete line radiation (bound-bound transitions), radiative recombination (free-bound transitions) and Bremsstrahlung (free-free transitions) produce a continuous X-UV spectrum.

Admitting that the multicharged ions of hot plasmas can work as X-ray emitters, special requirements have to be met in order the plasma may effectively amplify radiation. The characteristic feature of an amplified radiation is its relation intensity with the length covered in the emitting medium. G being the gain coefficient, L, the length, S the source function, the intensity reads (see figure 1):

\[ I = S(e^{GL} - 1) \]  

(1)
whereas at wavelengths where the medium is absorbing, with an absorption coefficient $\alpha$, one has:

$$I = S'(1 - e^{-\alpha L}) \quad (2)$$

Figure 1. Illustration of a two level radiative transition in a medium of finite length $L$. $N_i/g_i$'s are the so-called reduced populations, i.e. the total population of level $i$ divided by the statistical weight of the level.

both expressions reducing each other with the substitution:

$$G = -\alpha; S = -S' \quad (3)$$

Now, let $i$ and $j$ be the quantum numbers of two excited levels, the level $j$ lying above the level $i$; let $\nu$ be the $j-i$ transition frequency, $\Phi(\nu)$ the profile function of the transition, $g_i$ and $g_j$ the statistical weights. In thermal equilibrium at electron temperature $T_e$, the two level population densities, $N_i$ and $N_j$, are related by:

$$N_j/N_i = g_j/g_i \exp(-\hbar\nu/kT_e) \quad (4)$$

and the emitted radiation is re-absorbed inside the plasma, with an absorption coefficient:

$$\alpha = (\hbar\nu\lambda)\Phi(\nu)g_iB_{i,j}(N_j/g_j - N_i/g_i) \quad (5)$$

where $B_{i,j}$ is Einstein's absorption coefficient. If we know how to produce a population inversion in the plasma, we will have:

$$G = (\hbar\nu\lambda)\Phi(\nu)g_iB_{i,j}\Delta N \quad (6)$$

where $\Delta N$ is the inversion density:

$$\Delta N = (N_j/g_j - N_i/g_i) \quad (7)$$

In the 100 ev photon energy range, (6) leads to:

$$G \sim 10^{-15} \Delta N$$

Plasmas may produce population inversions either when a rapid change of temperature and density or if a strong energy flow in stationary conditions breaks down the thermodynamical equilibrium. For the theoretical prediction of population inversions as well as to make simulation of experiments, two computational models are generally used together. The first provides the time evolution of plasma density and temperature. The second uses these last as parameters in the system of rate equations which represents the atomic properties of the ions.

Now, given the size of laboratory plasmas and owing to the lack of high quality X-ray mirrors, to reach the gain-length value of 20 - 30 necessary for an X-ray laser, we must have:

$$G > 1 \text{ cm}^{-1}$$
Since the upper-level density is a maximum value of $\Delta N$ we can see that excited level density around $10^{15}\text{cm}^{-3}$ is necessary for population inversion to yield an effective amplification.

Such conditions can be satisfied in laser-produced plasmas. Figure 2 shows the principle of plasma production by cylindrical focusing of the laser beam onto target surface. This realizes a plasma "column" which has approximately constant properties along parallel-to-surface axis, while density and temperature vary in the perpendicular direction. The steepness of these variations depends on the exact target geometry and on the laser shot power. Density and temperature appropriate for lasing plasma are generally found between 50 $\mu$ and 500 $\mu$ from target surface.

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**Recombination lasers**

Early considered in hydrogen /3/, the recombination scheme has been extensively studied in H-like ions with a view of XUV amplification. Let us consider a plasma, initially completely ionized. Due to fast cooling, the plasma gets out of equilibrium. Three body recombination populates strongly the most excited levels of the recombined H-like ion. This is followed by collisional-radiative cascade to intermediate levels. At the same time, owing to free-electron low temperature, electron-ion collisions from the ground level can no longer balance the radiative decay of the lowest lying levels. As a consequence, transient population inversions occur between intermediate and lower levels.

In this respect, the group of Hull University has shown the adiabatic expansion of a freely expanding plasma to be able to produce a population inversion between 2-3 levels of H-like carbon at 182 $\text{A}$ wavelength./4/. A slightly different scheme, including plasma confinement in a strong sinusoidal magnetic field, has been developed by S. Suckewer et al. /5/. Besides the emission of radiation from ions having strong resonant lines does also contribute to plasma cooling. Lithium-like ions are of a special interest because intense is the line emission of the parent ions (He-like).

The case of aluminium is exemplified in figures 3 which give the variation of helium-like and lithium-like ion abundances during a 3 ns laser pulse (3a) and the $4f - 3d$ and $5f - 3d$ population inversions (3b) at fixed distance from the target, with corresponding $N_e$ and $T_e$. These inversions occur respectively at 154 $\text{A}$ and 105.7 $\text{A}$. /6/. One can see that the maximum of population inversion occurs about 3 ns after the laser pulse. This delay is typical of recombination mechanism of population inversion production.
Figure 3 a. Example of calculated ion abundances as a function of time in aluminium laser-produced plasma. Hatched area, laser pulse; solid curve, lithium-like ions; dashed curve, helium-like ions; dotted curve, electron temperature (maximum at 210 eV). The vertical arrow indicates the predicted moment of the 3d-5f population inversion outset.

Figure 3 b. Al^{10+} ion. Below: time-variation of 5f - 3d (solid curves) and 4f - 3d (dashed curves) population inversion densities in the space region of their maxima (0.35 mm from the target). Above: plasma electron density and temperature at the same position. Pulse duration (displayed in dashed area): 0.6 ns. a) Laser wavelength: 1.06 μ; flux density: 12.7 GW/cm. b) Laser wavelength: 0.53 μ; flux density: 12.7 GW/cm. c) Laser wavelength: 1.06 μ; flux density: 21.6 GW/cm.

Princeton experiment

In Princeton experiment (USA) the plasma is produced, along the axis of a 4-m radius spherical mirror, by a high power CO₂ laser, and it is confined by a sinusoidal magnetic field. The target may be either a gas or the edge of a hole in a solid disc, possibly completed by blades making the plasma more uniform /7/. This set up is represented in figure 4.

Figure 4. Device producing a magnetically confined laser-plasma at Princeton University. Amplified X-rays are detected by the axial monochromator.

In this system the column length is not measured as precisely as with cylindrical focusing optics. Moreover it cannot vary without changing other plasma parameters. Therefore the gain
Figure 5. On the left, enhancement of the 3-2 line of C^5+ from transverse to axial observation. The gain G.L deduced from this measurement is of 4.3. On the right, additional enhancement due to the XUV spherical mirror represented in figure 4.

is deduced from line intensities measured simultaneously in axial and transverse directions, the lasing transition intensity being normalized with the help of another line which must not to be affected by radiation trapping. Owing to magnetic confinement, the gain produced by plasma recombination does last a time longer than in other experiments.

A gain-length product, G.L, as large as 6.5 has been obtained at 182 Å (2-3 transition of H-like carbon). A further and very impressive proof of amplification is given by using an X-UV multilayer spherical mirror. The reflectivity of the mirror at 182 Å was measured to be 12 %, whereas the increasing of the line intensity along the axial direction, due to the mirror was about 100 % /8/. Figure 5 gives examples of the results.

Rutherford Laboratory experiment

One of the first, the group of Hull University (UK) has shown the adiabatic expansion of a freely expanding plasma to be able to produce a population inversion between 2-3 levels of H-like carbon provided that the expansion rate of the plasma is optimized by choosing an appropriate target geometry /4/.

Rutherford Laboratory has developed a laser-beam focusing system especially suitable for fibre target irradiation. In consists in a combination of focusing lens and off-axis spherical mirror which produces a line focus free of transverse aberration. Up to six laser beams can be brought round on the same thin fibre of 7-mm length. Various options in beam arrangement enable to choose among different lengths. A sketch of the interaction chamber can be seen in figure 6. This system has been successfully used for measuring the variation of intensity with length for the 3-2 line of C^5+ at 182 Å. The result is the curve shown in figure 7 which exhibits an exponential increasing with a gain coefficient about 4 cm⁻¹ /9/.

A similar result, with a gain coefficient of 3 cm⁻¹, has been reported for the 80;9 Å line of H-like fluor produced from a LiF coated fibre target /10/.
Figure 6. Sketch of the Rutherford Laboratory experiment, illustrating the focusing optics designed for thin fibre irradiation.

Figure 7. Line intensity of the 2-3 transition of carbon H-like ions (on the right) versus the length of plasma column, as compared to the 3-4 line (on the left).

Palaiseau experiment

Palaiseau experiment (F) uses the Greco "Interaction Laser-Matière" facilities. The laser beams have 90-mm diameter. Two of them are available for the present X-ray laser experiment. A focal line with a length up to 2 cm is produced on the surface of a massive target by a system of two crossed cylindrical lenses. The laser energy is about 200 J per pulse /6,11/.

The particular way of using here the property expressed by relation (1) consists in choosing a couple of two plasma lengths and to obtain a relatively extended gain (absorption) spectrum, at one go, from the recording of only two emission spectra in the same axial direction. If L and l are two plasma column lengths (at same density and temperature), $I_L$ and $I_l$ being the corresponding intensities at a given frequency, the gain coefficient for this frequency is obtained by solving the equation /12,13/:
Figure 8. Gain as a function of time for the 5f - 3d transition of Al$^{10+}$ measured for a 0.7 cm long plasma column. Laser-pulse half-maximum duration: 2.5 ns; flux density: 5x10$^{12}$ W/cm$^2$.

$$(e^G L - 1)/(e^G 1 - 1) = I_L/I_1$$

The plasma radiation is recorded with a streak camera providing a time-resolution about 100 picosecond. It can be connected with an OMA instead of the usual photographic camera. This provides directly digital data from the time-dependent plasma spectrum /14/.

A typical result achieved by using this procedure is presented in figure 8 which reproduces the time-dependence of the 5f-3d line of Al$^{10+}$ for a plasma column of length $L = X$ cm. The laser-pulse width is $X$ ns. The investigated plasma section is located at a distance $X = X$ mm from the target surface. In this case a gain with a peak value of $X$ cm$^{-1}$ is observed at the center of the line. The peak of gain displayed in figure 8 has been shown to occur in the expanding aluminum plasma about 5 ns after the top of the Nd-laser pulse.

The shortest wavelength for which a gain has been observed up to now in using recombining lithium-like ions is 65 Å. This is the wavelength of the 3d-5f transitions of S$^{13+}$. The value of the gain is near 1 cm$^{-1}$/15/.

For particular multicharged ions species, steady state population inversions are predicted by calculation as a result of the balance between collisional ground state excitation produced by plasma free electrons, on the one hand, and fast radiative decay of low lying levels, on the other hand /16,17/. The neon-like sequence is far the most extensively studied up to date. For neon-like ions the collisional excitation rate from the ground level is approximately the same for 1s$^2$ 2s$^2$ 2p$^5$ 3s and 1s$^2$ 2s$^2$ 2p$^5$ 3d levels. At the same time, owing to selection rules, the radiative decay from the 1s$^2$ 2s$^2$ 2p$^5$ 3p to the ground level is strictly forbidden, whereas it is quite large from the 1s$^2$ 2s$^2$ 2p$^5$ 3s level. Moreover there is a large rate of population transfer from 3d to 3p level by radiative and collisional cascades. As a consequence, population inversion can occur between the 3p and 3s levels provided that plasma free electron density is chosen in a suitable range. Rate equation calculations show that population inversions occur only in a rather narrow range of plasma electron density. At low density, there is no collisional pumping and, when the density becomes too large, collisional mixing between the levels leads to statistical equilibrium for populations. As an example, the gain calculated for neon-like strontium in the range of 160 Å wavelength is shown in figure (9) /18/.

Collisional pumping.

Ni-like sequence is also under active consideration. In this case inversions may appear on 4p-4d transitions /19/.
Livermore experiment.

Large gain coefficients have been measured at Livermore (USA), in 1985, with neon-like ions /20/. Livermore experiment uses two arms of the giant Nova laser, the beams of which have 74 cm of diameter. 2000-J laser energy, or more, can be deposited on the target with a pulse length about 0.5 ns. The target is a thin foil, for instance a 1500-A thick substrate of formvar coated with a 750-A layer of selenium. The sketch of the experimental set up is shown in figure 10.

A grazing incidence spectrograph, provided with a gated microchannel plate detector, records the soft X-ray signal in the direction of the axis of the plasma column. The evidence of amplification is given by the plot of the signal intensity versus the plasma length. The gain coefficient is found to be near 5 cm⁻¹. Figure 11 shows a result obtained with a selenium plasma of 4 cm length. The gain-length product in this experiment is as large as 16 /21/.

Since the first experiments, a considerable progress has been made at Livermore on the way of a true X-ray laser. A successful attempt of lasing inside a cavity, which is made of a spherical multilayer mirror and a semitransparent multilayer thin plate /22/, has been recently performed /23/. In addition, the Ni-like sequency has been proved to yield amplification at short wavelengths. A gain about 1 cm⁻¹ has been observed at 66 A with europium /24/.
Various attempts to realize collisional pumping with more economic means than in Livermore experiment have been made /25,26/. A recent result obtained at Limel (F) is presented in figure (12). It shows the large increasing of lines at 232 A and 236 A according to plasma length. The lines belong to the spectrum of Ne-like germanium. The target was a thin metal layer on plastic foil. The irradiation is made by six beams of the laser Octal, delivering $7 \times 10^{12}$ W/cm$^2$ on the focal line on each side of the target /25/.

**CEL experiment**

Besides recombining plasmas and collisional excitation, a very different approach is still to be mentioned. It consists in looking for population inversions created by multiphoton absorption of KrF-laser radiation /27/. This type of laser can deliver powerful gusts of five-ev photons. Starting from the 4s$^2$2p$^5$ ground state of Kr-like Cd$^{12+}$ ions, for instance, one could populate the 4s4p$^5$5p state. Then gain could take place on either the 5p-4d or 4p-4s transitions. Pumping must be done on a picosecond time scale in order to avoid the radiative decay of the upper to the ground state. Laser intensity of the order of $10^{15}$ W/cm$^2$ is required.
Conclusion

Works described in the previous sections show that hot plasmas of sufficient density are able to amplify soft X-rays. As regards the comparison of X-ray lasers with other sources, a very large enhancement will occur in their brightness. Assuming "realistic" experimental conditions in which the observed emitting area should be $10^{-3}$ cm$^2$, the solid angle at the entrance of the measurement device being $10^{-6}$ steradian, one finds approximately the same number of $1 - 5 \times 10^4$ photons per pulse for a line at 100 ev ( =120 A) emitted by laser plasma and tokamak. But the pulse duration is $10^8$ times shorter for laser than for tokamak, leading to the same ratio between their brightnesses, to laser advantage. In a similar way, typical photon number and brightness are compared in table 1 for various sources, in letting in a thermal line broadening of 0.02% for plasmas and using the same band width for other sources. Although we did not explain yet how will work an X-ray laser, it is interesting to compare its predicted capability to other devices in the same photon energy range. The estimate displayed in table 1 is made out in assuming a population inversion density of $10^{15}$ cm$^{-3}$ in a 10 cm long amplifying plasma column.

<table>
<thead>
<tr>
<th>Source</th>
<th>Pulse duration(sec)</th>
<th>Photons per pulse</th>
<th>Brightness (a.u.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tokamak plasma</td>
<td>$10^{-1}$</td>
<td>$1 - 5 \times 10^4$</td>
<td>1</td>
</tr>
<tr>
<td>Laser plasma</td>
<td>$10^{-9}$</td>
<td>$1 - 5 \times 10^4$</td>
<td>$10^8$</td>
</tr>
<tr>
<td>Storage ring</td>
<td>$2 \times 10^{-10}$</td>
<td>$2 \times 10^3$</td>
<td>$10^8$</td>
</tr>
<tr>
<td>Undulator</td>
<td>$2 \times 10^{-10}$</td>
<td>$2 \times 10^8$</td>
<td>$10^8$</td>
</tr>
<tr>
<td>Soft X-ray laser</td>
<td>$5 \times 10^{-10}$</td>
<td>$10^{12}$</td>
<td>$2 \times 10^{16}$</td>
</tr>
</tbody>
</table>

From now onwards, systems involving hydrogenic, lithium-like and neon-like multicharged ions can be used for developing amplifiers. A propitious circumstance is these systems to provide fairly large gain coefficients of the order of $1$ cm$^{-1}$ - $5$ cm$^{-1}$. Given the moderate reflectivity which can be expected from soft X-ray mirrors, large gain coefficient is indeed an essential condition for achieving X-ray lasers. A great deal of work is still to be done for improving the control of large gain production.

To succeed in overtaking the saturation threshold, above which there are approximately as much photons as inverted atoms in the active medium, the using of interferential mirrors has to be extended and, on the other hand, it is necessary to increase the length of plasma column. In keeping constant the heating flux density on the target, increasing length needs more energy. At Palaiseau we will use five laser beams for producing 6-cm long plasmas with a few tens of GW/cm.

Difficulties for achieving large gain in long plasma can appear from several directions. Defects in laser-target coupling may cause an erratic spatial distribution of gain coefficient, preventing amplification to develop completely along the axis. Second, the density gradient perpendicular to the target surface does produce refraction, which may entail a similar consequence in bending the trajectory of the radiation. New target designs, minimizing this gradient, are thus necessary, especially for systems working at large density. Finally, increasing of plasma size tends to increase also the radiation trapping for non-lasing lines. In recombination systems this may reduce the population inversion by photoexcitation from low-lying levels. New constraints for target design result from this, with a view to make transverse escape of radiation as free as possible.

Shorter wavelengths are under active consideration for future X-ray laser experiments. The schemes described above can be extended to ions of higher Z provided that plasma temperature is large enough. The wavelength-to-temperature scaling differs considerably according to the population inversion scheme. This is shown in terms of density flux deposited on target surface in figure 13 where the points below 50 A and the lower point of molybden are calculated.
In order to decrease the lasing wavelength more energy must be spent for producing the plasma. This figure shows how the density power onto the target does scale with the wavelength for the collisional schemes (Ne-like and Ni-like ions) and the recombination schemes (H-like and Li-like ions). The dashed vertical lines delimit the "water-window" which is of a great interest for biological applications.

Lithium-like ion plasma allows an especially economical extension to short wavelengths because, in this case, lasing wavelength and plasma temperature have approximately the same $Z^2$-scaling. The figure displays the "water-window" which is a wavelength range of great interest for biological applications. Inside this window, radiation is absorbed by the K-edge of carbon atoms of organic molecules but not by the oxygen K-edge of water molecules. Thus using an X-ray laser in the water-window would make possible, for instance, holography of living samples.

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