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# Quantitative study of 10 Hz operation of a soft x-ray laser — energy stability and target considerations

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**Abstract:** A soft x-ray laser from Ni-like Mo, pumped in grazing incidence (GRIP), is analyzed with regard to high repetition rate operation. Reliable lasing is obtained, but with significant energy fluctuations attributed mainly to beam pointing jitter from the pump laser. Two modes of operation are compared: continuously moving target and stationary target. With a moving target the soft X-ray output is constant on average, whereas the repeated use of the same target position leads to a pulse energy which increases for several tens of shots. This effect might be caused by improved guiding of the pump laser in the formed groove and the removal, through laser ablation, of the oxide layer on the target surface.

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**OCIS codes:** (140.7240) UV, XUV, and X-ray lasers; (340.7440) X-ray imaging.

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## 1. Introduction

Among the many applications for light in the soft X-ray region (wavelengths up to a few tens of nm) are holography, interferometry, lithography and materials processing. Currently, this type of radiation is generated by e.g. synchrotrons [1] or by upconverting a longer wavelength laser using high order harmonic generation (HHG) [2]. These methods have their specific advantages and drawbacks. Synchrotron radiation has tunable wavelength, high average power and brightness, and very high repetition rate ( $\sim 100$  MHz), but has low coherence and requires large, costly facilities. HHG from visible and near infrared (IR) lasers have excellent coherence and can be generated with, in this context, low-cost table-top laser systems operating typically at 10 Hz or 1 kHz. However, they suffer from relatively low pulse energies.

Soft X-ray lasers (SXRL) present an interesting alternative to synchrotrons and harmonic generation. They offer the possibility of high pulse energy and peak brightness, together with good temporal coherence [3]. The pulse duration is typically a few picoseconds [4], i.e. in a range between HHG (few tens of femtoseconds or even attoseconds) and synchrotrons ( $\sim 100$  ps). The SXRL generating process is based on population inversion of an electronic transition in the selected ion species, that consequently completely determines the wavelength of the laser. Ever since the first SXRL was demonstrated about 20 years ago [5], they have been associated with high energy pump lasers. Inherent to such systems are very low repetition rate and large cost. But, the amount of pump energy needed for a saturated X-ray laser has decreased gradually over the years. Notable advances are the introduction of transient collisional pumping [6], and more recently grazing incidence pumping (or GRIP) [7]. Now, a saturated soft X-ray laser can be produced with less than one joule of pump energy, making high repetition rate operation (see e.g. [8, 9]) possible at relatively low-cost, university scale facilities. This development makes the SXRL a promising tool for a wide range of applications, and the field of XRL studies has therefore received increased interest world-wide.

The GRIP technique is a version of the transient collisional pumping scheme, where the plasma formation and the actual pumping of the laser transition is separated into two pulses. First, a long (hundreds of ps) IR laser pulse hits the target and creates a plasma which is allowed to expand and form a smooth density gradient. After a given time delay, the second, short (a few ps), pump pulse is incident on the plasma at a grazing angle,  $\phi$ , and heats free electrons through inverse bremsstrahlung absorption. As opposed to the normal incidence pumping in the original transient collisional method, the pump is refracted by the plasma density gradient and changes

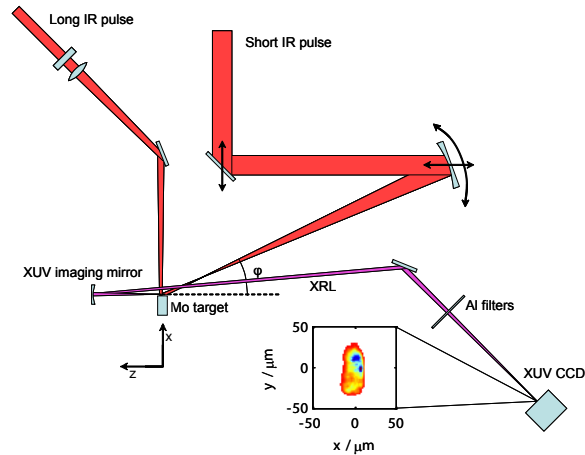


Fig. 1. Overview of the experiment setup. The XRL near field imaging diagnostic is indicated. Inset is an image of a typical near field SXRL intensity distribution.

direction at a plasma density that depends on the angle of incidence. In the turning point region, laser energy is efficiently transferred to the plasma. Advantages of the GRIP scheme, compared to normal incidence, are less refraction of the SXRL beam due to the smooth plasma density gradient, pump energy deposition localized to the gain region and increased path length of the pump laser in the gain medium.

In a recent study, we have investigated various aspects of optimizing grazing incidence pumping, such as grazing angle  $\varphi$ , delay and energy ratio between the long and short pulses [10]. Single shot soft X-ray energies of up to  $3\mu\text{J}$  were measured. This is comparable to the  $2.3\mu\text{J}$  per shot obtained by Li *et al.* [11] for a saturated Ni-like molybdenum X-ray laser using a much larger 10J driving laser. We report here findings related to 10Hz operation during the experiment described in [10] and in particular on energy stability and target considerations.

## 2. Experimental

The driving laser is the 10Hz multi-TW laser at the Lund Laser Centre. It is a Ti:sapphire CPA [12] system, with 800nm central wavelength, delivering pulses with up to 1.6J before compression. When fully recompressed, the pulse length is 35 fs FWHM. However, the GRIP technique requires a longer,  $\sim 1$  ps, duration for the grazing incidence pump pulse. This was accomplished by moving a grating in the compressor. Part of the uncompressed ( $\sim 300$ ps) beam was split off after the last amplifier and used to preform the plasma medium. A spherical / cylindrical lens package was used to focus the long pulse beam to a line,  $70\mu\text{m} \times 5$  mm FWHM. Refractive optics is not an option for the short pump pulse, due to non-linear effects induced at high intensity. Instead, a spherical mirror was used off-axis to produce an astigmatic focal line. The grazing angle on target, which is twice the angle on the spherical mirror, varied between  $15^\circ$  to  $21^\circ$ . The line focus was about  $40\mu\text{m}$  wide and 6 to 8mm long depending on the angle. The target was a 5 cm long and 4mm wide slab of molybdenum, polished with standard mechanical grinding. The selected lasing transition was  $4d^1S_0 - 4p^1P_1$  in the Ni-like ion  $\text{Mo}^{14+}$  at 18.9 nm. For this study, the energy content in the driving laser pulses were 0.5J in the long pulse and 0.5J in the 5 ps short pulse. The delay between them was set to 500ps. The optimum GRIP angle was found to be  $19^\circ$ . An additional prepulse, containing 14% of the total long pulse energy, preceded the main pulse by 2.3 ns to improve the plasma conditions.

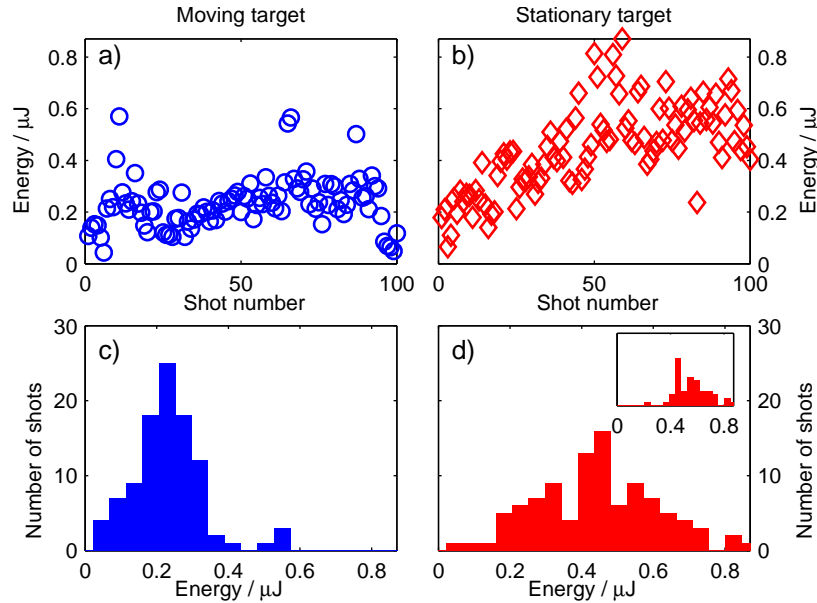


Fig. 2. X-ray laser output energy time series and histogram for moving target (parts a and c) and stationary target (parts b and d). Inset in d) is the histogram for the last half of the shot sequence. Mean energy is  $0.22 \mu\text{J}$  and  $0.42 \mu\text{J}$  for moving and stationary targets respectively.

The main diagnostic instrument was a high-resolution near field imaging device. It imaged the soft X-ray laser energy distribution at the exit of the gain region, by means of a curved X-ray mirror, onto a CCD chip. The pixel resolution was  $1.7 \mu\text{m}$  at the object plane, corresponding to a magnification factor of about 7. Figure 1 shows a schematic of the setup. Aluminium filters of different thicknesses were used to attenuate the SXRL beam and to reduce the plasma background emission. To achieve 10 Hz frame rate, the camera was set to capture only a  $46 \times 41$  pixel region of interest. The small area combined with the moving source position for a stationary target limited the number of shots to about 100 in a single run.

### 3. Results

10 Hz operation of the X-ray laser is tested in two different ways. First the target is moved at a constant rate of  $0.4 \text{ mm/s}$ , corresponding to the width of the focal spot between shots, to supply a fresh target surface for every shot. Then the target is stopped, allowing the pump laser to dig into the bulk of the target. This is done in immediate connection in time during a total of 20 s (200 shots), to minimize any effect of laser drift. Time series of the integrated energy in every shot are shown in Fig. 2. The moving target has a stable output level (there are no missed shots) with an average SXRL pulse energy of  $0.22 \mu\text{J}$  and a standard deviation of  $0.09 \mu\text{J}$ , i.e. 41% of the mean. When the target is stopped, the pulse energy ramps up during approximately 50 shots, leveling off at mean energy of  $0.53 \mu\text{J}$  and a slightly higher standard deviation  $0.11 \mu\text{J}$ . The relative standard deviation is thus lower, at 21% of the mean. Additional tests indicate that the higher output level for a stationary target can be sustained for several hundred shots.

The RMS size of the near field emission region is roughly  $7 \mu\text{m}$  in both vertical and horizontal directions, see Fig. 3. The shot-to-shot fluctuations are much larger in the vertical direction than in the horizontal. There is, however, no significant change in source size for the two cases

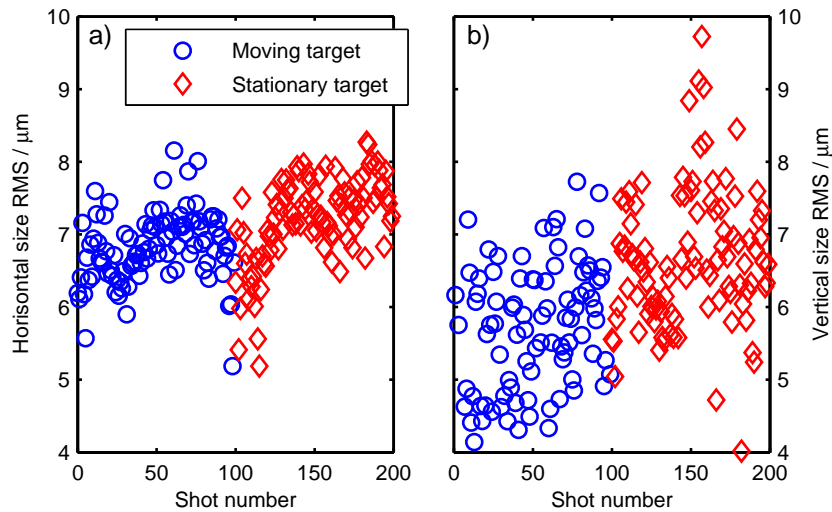


Fig. 3. Size of the near field distribution of the soft X-ray laser for both moving and stationary targets. In a), the horizontal size (perpendicular to the target surface) is plotted and b) shows the vertical size (tangential to the target surface).

of moving or stationary target.

Figure 4 shows the distribution of the distance of the X-ray laser near field centroid positions to the target surface for the two shot series. The SXRL emission centroids with a moving target are clustered in a region roughly  $10 \times 15 \mu\text{m}$  in size, corresponding to the beam pointing of the driving laser. When the target is stopped, the centroid position starts moving steadily in toward the original target surface, at a rate of  $\sim 0.22 \mu\text{m}/\text{shot}$ .

#### 4. Discussion

The shot-to-shot fluctuations in output energy can partly be attributed to fluctuations in the driving laser itself, in terms of both energy per pulse and beam pointing. The IR laser has shot-to-shot energy fluctuations of the order of 10%. A graph showing SXRL output energy as a function of short pulse pump energy is plotted in Fig. 5(a). The laser parameters used to obtain this graph were slightly different from the ones in the rest of the paper, but the general shape of the curve is independent of these differences. Clearly, at the 500 mJ pump energy used in the present experiment, a 10% variation in laser energy results in  $\sim 10\%$  variation in the SXRL pulse energy.

Another likely cause of energy fluctuations is beam pointing jitter of the pump lasers. The two line foci,  $40 \mu\text{m}$  and  $70 \mu\text{m}$  wide respectively, must overlap perfectly to ensure optimal X-ray lasing. A scan where the long pulse line focus was moved relative to the short one is found in Fig. 5(b). The width of the SXRL peak is only  $\sim 30\%$  of the long pulse focal line. This is because the lasing ions are produced only in the central, high-intensity, part of the focus, due to the gaussian intensity profile of the pump laser [13]. With an estimated  $15 \mu\text{m}$  shot-to-shot fluctuation, due to mechanical vibrations, of the relative positions of the focal spots, Fig. 5(b) indicates that there will be significant variations in output energy, consistent with our observations.

The target surface conditions are also believed to have a significant effect on the observed behavior for both stationary and moving target operation. After the experiment, the target slab

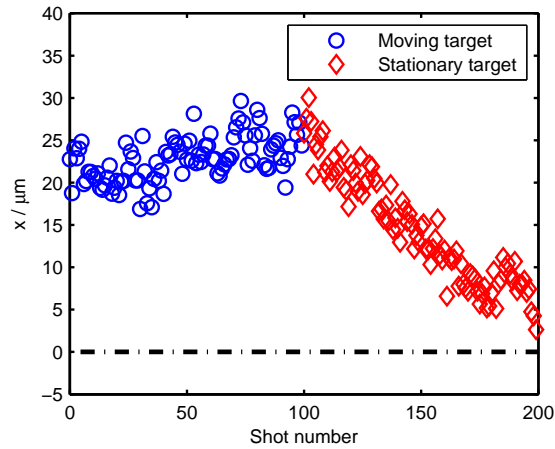


Fig. 4. Distance between the centroid of the X-ray laser near field and the target surface for both moving and stationary target. The original target surface is indicated with a dashed-dotted line. With a moving target, the gain region is hovering  $23\ \mu\text{m}$  from the target (a situation corresponding to single shot operation). When the target is stopped, the pump laser ablates the target, causing the gain region to move inward at a constant rate.

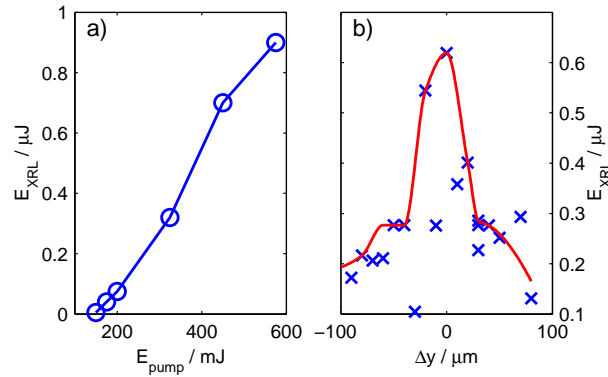


Fig. 5. The output X-ray laser energy versus short-pulse pump energy is plotted in a). This particular data set is for  $\varphi = 17^\circ$  and 600 ps delay between long and short pulse. In b), the sensitivity of the X-ray output to the overlap between the two line foci is shown. The red line outlines the envelope of the shots. FWHM of the peak is approximately  $30\ \mu\text{m}$ .



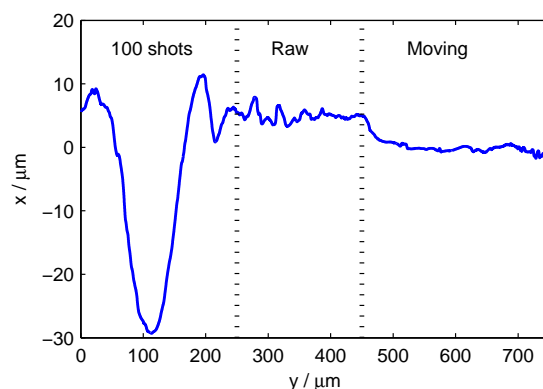


Fig. 6. Target profile in the plane normal to the SXRL axis, as measured with a surface profilometer. Three regions are highlighted: irradiated stationary target, raw target and irradiated moving target. The reduced roughness of the target surface after a single laser exposure is clearly seen in the moving target region. Firing 100 shots on the same target position produces a deep crater with material thrown up to the sides.

has been analyzed with a surface profilometer. The results are shown in Fig. 6. The unexposed raw surface roughness is initially  $1\ \mu\text{m}$ , and after one shot it is smoothed to  $\sim 0.5\ \mu\text{m}$  (such as in the moving target case). This could account for the larger shot-to-shot variations for a moving target, since it has been shown that the laser–plasma interaction is strongly dependent on the surface quality on a microscopic scale [14].

The higher output energy for the stationary target could also to some extent be explained by the reduced surface roughness. A smoother target surface leads to the creation of a more homogeneous plasma and a better interaction with the pump laser. However, this smoothing should only occur during the first few shots, and not provide additional improvement for the next several tens of shots. The groove produced during the tests with a stationary target improves the coupling between the plasma and the pump laser [15, 16], and can thus lead to the gradual increase in SXRL energy over the first 50 shots. The depth of the groove is  $30\ \mu\text{m}$ , i.e. the IR laser is eating into the target bulk at rate of  $0.3\ \mu\text{m}/\text{shot}$ . This is in fair agreement with the observed movement of the source position. As evident from Fig. 6, the material is to a large extent thrown up to the sides, forming walls on either side of the groove, providing additional guiding of the pump laser.

The presence of an oxide layer ( $\text{MoO}_2$ ) [17] on the target surface is an additional reason for the lower output level observed with the moving target, since the oxide leads to a lower density of lasing ions and hence a lower gain. The oxide layer is very thin [18], and should be removed by the first few laser shots. This has been confirmed with a spectrometer, where  $\text{O}^{5+}$  from plasma emission and SXRL lines were simultaneously recorded. After five shots, the  $\text{O}^{5+}$  line almost disappears, whereas the SXRL intensity still increases.

## 5. Conclusions

High repetition rate operation of a soft X-ray laser has been characterized with quantitative precision by means of real time acquisition of near-field images from the exit aperture of a soft X-ray laser pumped in GRIP configuration. This diagnostic allows for energy and 2D-spatial characterization of the X-ray emission as a function of shot number over series of a few hundreds shots. We confirm the results obtained by Weith *et al.* [9], concerning the level of



energy fluctuations for a Ni-like molybdenum X-ray laser ( $\sim 20\text{--}40\%$ ).

Moreover, we compared two different types of operation, moving or stationary target, which exhibit distinct behaviors. With the moving target, the pump laser is always incident on a fresh target and we observe a significant reduction of the output energy and an increase of the fluctuations. This is partly attributed to the existence of a thin layer of molybdenum oxide, which is supported by spectroscopic observations. The roughness of the target also plays an important role by degrading the plasma homogeneity. Both these effects could be removed in the near future by irradiating the target with a low intensity laser before an X-ray laser shot.

With the stationary target, we observe a progressive and significant increase of the output energy over the first 50 shots, accompanied by a constant drift of the source position toward the target direction at a rate of  $0.3\ \mu\text{m}$  per shot. This behavior is attributed to the formation of a groove that could affect laser absorption and/or plasma expansion. It seems thus more advantageous to operate the Mo X-ray laser with a stationary target, since the improved target surface conditions leads to an increased output energy. However in application experiments, the constant drift of the source towards the target by a few  $10\ \mu\text{m}$ , which is not a severe problem if the source is used in free propagation, could become problematic if the source is used in imagery.

Finally we show that the fluctuations of the pump laser, in terms of energy and beam pointing, contribute to the shot-to-shot variations in the SXRL energy. Possible solutions to improve the stability of the SXRL include: increasing the pump energy to reach saturated amplification where the sensitivity to pump energy fluctuations is reduced, increasing the width of the plasma to reduce sensitivity to pointing variations, and installing vibration decoupling in the pump laser to improve the pointing stability. In conclusion, thanks to the above study and with simple and realistic improvements, the 10 Hz operation of soft X-ray laser becomes possible for applications in imagery and irradiation having the same stability requirements than for any usual infrared laser facility.

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