

# Characterization of a Seeded Optical-Field Ionized Collisional Soft X-Ray Laser

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**Abstract.** By seeding an optical-field-ionized population inverted plasma amplifier with the 25th harmonic of an infrared laser we have produced a compact, diffraction- limited and Fourier-limited laser beam in the soft x-ray spectral range. This laser beam is emitted within a cone of 0.7 mrad at a repetition rate of 10 Hz at a central wavelength of 32.8 nm. The beam exhibits a regular Gaussian spatial profile, and wavefront distortions smaller than  $\lambda/17$ . The measured coherence time of 5.5 ps is equal to the duration of the lifetime of the amplifying plasma which shows that this source has reached the Fourier limit.

## 1 Introduction

The high scientific activity related to the development of ultrafast, coherent soft x-ray sources is motivated by the large number of novel applications such as high-resolution microscopy, lithography, interferometry and holography that can be realized with such sources. Recently, fourth generation synchrotron sources have been designed to provide highly coherent, bright, soft x-ray beams, able to open up new opportunities in science (1). In parallel, dramatic advances in ultra-short pulse laser technology have made it possible to generate compact, high repetition rate, coherent soft x-ray sources. The most advanced sources are based on high-order harmonic generation (HHG) and plasma soft x-ray lasers (SXRL). HHG are produced by focussing a high intensity femtosecond laser pulse into a gas medium. Odd harmonics of the fundamental laser frequency (i.e.  $3\omega$ ,  $5\omega$ , ...) are produced in a directional, collimated beam with photon energy that can be extended up to 1 keV (2). HHG driven by current optical lasers exhibit high spatial coherence (3) with an average energy of a few hundreds of nanojoules at 30 nm (4-5). In contrast to HHG, SXRL provide much higher output energy per pulse and narrower linewidth (6).

The first laboratory SXRL at 20 nm was demonstrated nearly 20 years ago (7), and required a few kilojoules of laser energy for pumping. However, recent spectacular progress in this field has led to the development of a large variety of compact SXRL systems providing high energy, monochromatic coherent radiation down to 10 nm. The amplifying medium is a hot, dense, highly charged plasma column generated by the interaction of an intense infrared laser pulse with a solid or gas target (8-9) or by a high density capillary discharge in a rare gas (10).

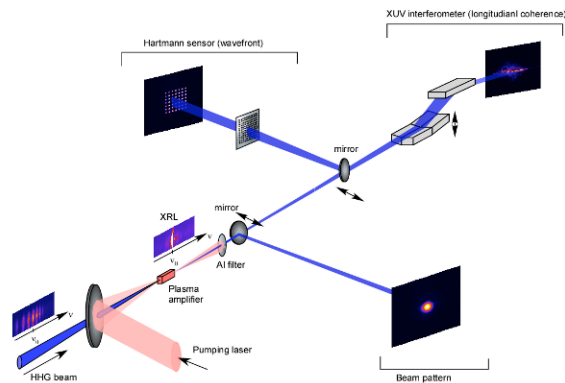
Until now, in all the SXRL schemes operating at saturation, population inversion between the levels of the lasing ion is induced by electron collisional excitation, leading to high gain value at short wavelength. The short lifetime of the gain and the absence of high reflectivity soft x-ray optics, make the use of this SXRL amplifier in a complete laser cavity impossible. For this reason, SXRL emission generally results from the single-pass amplification of spontaneous emission. As a consequence, SXRL radiation is characterized by rather poor optical qualities in terms of beam profile homogeneity, spatial coherence, source size regularity and wavefront distortion, which prevents the use of SXRL for applications that require a highly intense, coherent, soft x-ray photon flux in a sub-micron spot size.

To fully explore the potential of plasma-based SXRL sources, improving their spatial beam quality is a crucial bottleneck to be overcome. The solution we have chosen to explore consists in seeding a soft x-ray amplifier with a HHG beam (11). This approach is a direct analogy of the “oscillator-amplifier” concept commonly used for infrared laser system, applied to the soft x-ray range. Here the HHG beam plays the role of an oscillator which is injected and amplified while propagating through a population-inverted SXRL plasma column amplifier. This emerging scheme (12-13) offers a prospect for compact SXRL chain combining both the high energy extracted from the SXRL amplifier as well as the high optical quality of the HHG seed.

Here we report an experimental demonstration of a compact, diffraction- and Fourier-transform-limited laser beam in the soft x-ray range. The beam was emitted by 32.8 nm Optical Field Ionised (OFI) SXRL amplifier seeded by a HHG beam. Thanks to the spatial filtering of the seed beam by the plasma amplifier, the 32.8 nm laser beam exhibits a regular Gaussian intensity profile with a divergence of 0.7 mrad. The wavefront distortions were measured to be smaller than  $\lambda/17$  (1.9 nm) which demonstrates that this 32.8 nm laser beam is diffraction-limited. The temporal characterization shows that the measured coherence time is equal to the duration of the gain life time, i.e. 5.5 ps, which is the upper limit of the duration of 32.8 nm radiation. This consequently proves that this 32.8 nm seeded laser is Fourier-limited in the spectro-temporal domain. The laser line exhibits a Gaussian spectral profile with a full-width-at-half-maximum (FWHM) of only 3.61 mÅ.

## 2 Experimental set up

The experimental set-up is schematically illustrated in Figure 1. The experiment was performed using a 10 Hz, multi-terawatt Ti:sapphire laser system providing two independent 34 fs laser beams at a central wavelength of 815 nm. A first laser beam, containing about 10 mJ, was used to generate the seed HHG beam inside a 7 mm long gas cell filled with 30 mbar of Ar. We paid special attention to the 25th harmonic of the infrared laser which can be closely matched to the wavelength of the lasing transition of the SXRL amplifier. A grazing incidence toroidal mirror was implemented to image the output of the HHG source with a magnification of 1.5 at the entrance of the amplifier cell. The HHG seed spot was measured to be astigmatic with dimensions of about  $50 \times 100 \mu\text{m}$  (at  $1/e^2$ ). A second beam delivers  $\sim 600$  mJ on target and was used to create the OFI amplifying plasma column which drives the  $3d94d(1S_0) \rightarrow 3d94p(1P_1)$  transition of the  $\text{Kr}^{8+}$  ion at 32.8 nm (14). The pump beam was circularly polarized and focused by a 1 m focal length spherical mirror to a spot diameter of  $38 \mu\text{m}$  (at  $1/e^2$ ) inside a 7.5 mm long gas cell filled with Kr.

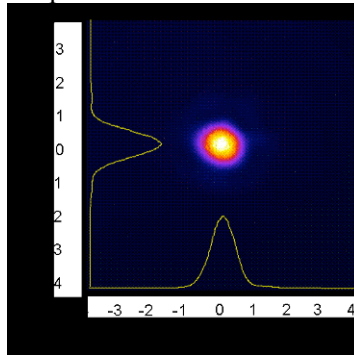


**Fig. 1** Schematic description of the experimental arrangement.

Under these conditions, the amplifier exhibits remarkable properties, well suited for the amplification of a seed pulse. The amplifier has a circular aperture and the gain value does not significantly vary along and across the gain column (15). The transverse density gradient is weak enough to prevent the seed beam from serious refraction (i.e. deterioration) during amplification. Previous simulations and experimental observations indicated that the amplifier has rather sharp edges which is an important criterium to ensure the efficient selection of the amplified seed radiation, i.e. spatial filtering of the injected seed beam (16).

### 3 Experimental results

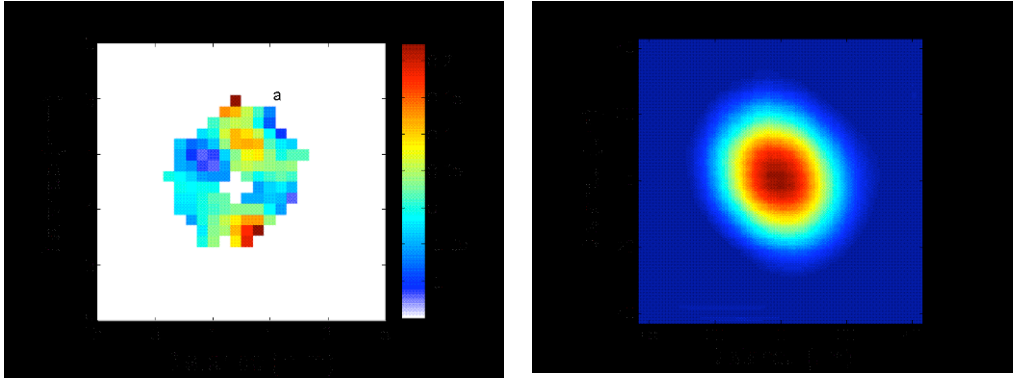
First, the 32.8 nm emission from the amplifier was optimized by varying its length, gas density, laser beam polarization, and focussing position. The most effective configuration was obtained for a pressure of 30 mbar, a cell length of 7.5 mm, and circular polarization. Second, the spatial and temporal overlapping between the HHG and SXRL beams was adjusted to produce the largest amplification of the seed radiation. The amplification started at a time delay between the SXRL plasma creation and HHG injection of 2 ps, reached a maximum ( $\times 180$ ) at 3 ps, and lasted up to about 8 ps. This direct measurement of the time evolution of the gain of a SXRL amplifier (17) indicates that the actual duration of the amplified seeded pulse does not exceed 6 ps. Then, using several online diagnostics schematically shown in Figure 1, we characterized the intensity distribution, the wavefront, and the transverse and longitudinal coherence of the amplified 32.8 nm laser beam.



**Fig. 2** Spatial profile of the seeded 32.8 nm laser beam.

The spatial profile of the seeded 32.8 nm laser was measured by placing a removable  $45^\circ$ , soft X-ray mirror made of Mo/B4C/Si tri-layers in the beam path, which redirected the beam towards a cooled, thin, back-illuminated charge-coupled device (CCD) camera. A 300-nm-thick aluminium filter was used to block the infrared beams. Figure 2 shows a typical profile of the generated seeded laser beam. The intense, monochromatic radiation is emitted in the direction of the HHG seed, being confined within a 0.7 mrad cone (FWHM). The spatial beam distribution is nearly perfectly Gaussian. Note that such a high beam quality has never been observed for other plasma-based SXRLs. In general, SXRLs exhibit highly contrasted intensity modulations (or speckles) forming a complex and irregular pattern (18), which is a direct consequence of low spatial coherence combined with the high temporal coherence of the amplified spontaneous emission (ASE) radiation (19). Here we take advantage of the fact that the HHG seed is partially coherent to avoid

intensity modulations in the beam pattern. In addition, the circular aperture of the gas amplifier permitted to tailor the intensity distribution of the 32.8 nm radiation, resulting in the Gaussian beam profile.

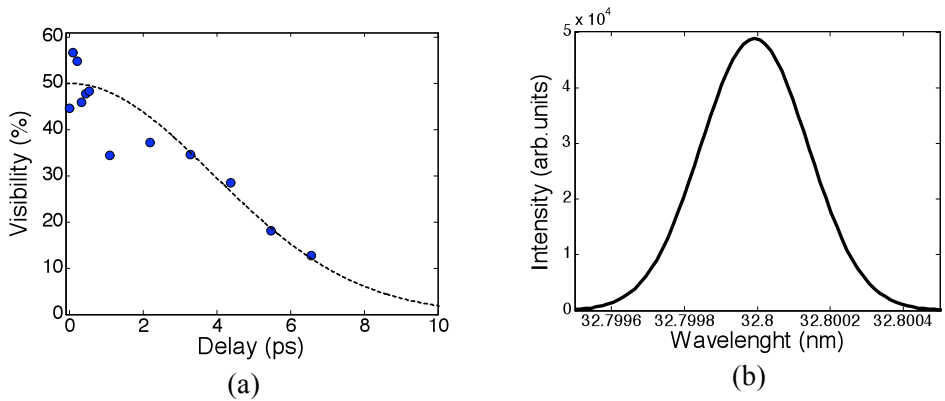


**Fig. 3** (a) Measured wavefront and (b) reconstructed source size of the seeded 32.8 nm laser beam.

A crucial parameter to evaluate laser beam quality is the wavefront. The wavefront of the seeded 32.8 nm laser was measured using a soft x-ray Hartmann sensor with an accuracy better than  $\lambda/20$  at 32.8 nm. In the Hartmann wavefront analysis a beam passes through a hole array and is projected onto a CCD camera that detects the beamlet sampled by each hole. The position of each individual spot centroid is measured and compared with a reference position. This enables the wavefront's local slope to be measured at a large number of points within the beam from which the wavefront can be reconstructed (20). Earlier investigations showed that neon-like Ar capillary discharge-driven SXRL at 46.9 nm exhibits wavefront distortion of  $3 \times \lambda_{\text{SXRL}}$ , which was the only measurement performed for plasma-based SXRL source (21) so far. Figure 3(a) shows the reconstruction of the wavefront of the 32.8 nm seeded laser. The amplitude of the wavefront defects never exceeds  $0.058 \times \lambda_{\text{SXRL}}$  (root-mean-square) which corresponds to beam wavefront distortions less than  $\lambda/17$  (1.9 nm). According to the Marechal criterion, which states that a system is regarded as well corrected if the wavefront distortion does not exceed  $\lambda/14$  (22), the generated seeded laser beam is clearly diffraction-limited. Note that the wavefront of the HHG seed beam was about  $\lambda/3$  due to the severe astigmatism introduced by the toroidal mirror. The dramatic improvement of the wavefront of the 32.8 nm laser is a direct consequence of the spatial filtering by the amplifier. This measurement also made possible the reconstruction of the intensity distribution of the 32.8 nm laser beam at the exit of the amplifying plasma column which results from the convolution of the wavefront shape with the intensity distribution of the beam. As shown in Figure 3(b), the source shape is Gaussian with a diameter of 50

$\mu\text{m}$  (at  $1/e^2$ ) which is in reasonable agreement with previous simulations predicting that the diameter of the amplifying zone of the plasma is about  $60 \mu\text{m}$  (16).

Due to the intrinsically narrow ion line width and the gain narrowing effect, SXRLs represent the most monochromatic sources in this spectral range. The spectral shape and line width of the  $32.8 \text{ nm}$  seeded laser has been inferred from the experimental measurement of the longitudinal coherence (19) by means of a wavefront division interferometer (Fig. 1). The SXRL beam was directed to the dihedron pair with a grazing incidence angle. After reflection on the dihedrons the incident beam was separated in two parts that converge and finally overlap in the far field, yielding interference fringes. As one of the dihedrons can be accurately translated in the vertical direction, a controlled delay (i.e. path difference) between the two interfering beams can be introduced. By following the decrease in fringe visibility with increasing delay, it is possible to reconstruct the spectral profile of the radiation with a resolution power better than  $2 \times 10^5$ .



**Fig. 4** (a) Variation of the measured fringe visibility as a function of the path difference. A decreasing Gaussian function was used to fit the data. (b) Reconstructed profile of the  $32.8 \text{ nm}$  laser line.

Figure 4(a) shows the measured fringe visibility as a function of the path difference. The experimental data were fitted with a Gaussian decreasing function from which the coherence time of  $5.5 \text{ ps}$ , defined as the path difference that decreases the maximum visibility by a factor  $1/e$ , was inferred. The fact that this coherence time matches the measured life time of the SXRL gain medium, clearly demonstrates that the  $32.8 \text{ nm}$  seeded laser is longitudinally fully coherent. Assuming a Gaussian spectral profile, we have inferred a SXRL line width of  $\Delta\nu = 1.1 \times 10^{11} \text{ Hz}$ , corresponding to  $\Delta\lambda = 3.61 \text{ m\AA}$ . Simulations carried out using the COFIXE code predict an ion temperature of  $T_i = 6 \text{ eV}$  (23). Under these conditions the Doppler-broadening of the line is

equivalent to 6.8 mÅ. Other calculations performed with the PPP code (24) showed a negligible effect of Stark broadening. The homogeneous broadening due to electron collisions has been calculated within the atomic model described in (16), leading to a value of 5 mÅ at 30 mbar. The effect of the amplification on the line shape has been calculated using the model described by J. Koch et al. (25), and assuming a uniform plasma column. For the amplification factor equal to the experimental value, the shape of the SXRL line is Gaussian with FWHM of 3.2 mÅ, in good agreement with the experimental results

## 4 Conclusion

By seeding of a laser created plasma amplifier we have demonstrated that it is possible to generate an intense soft x-ray beam having all the fundamental properties of common visible/IR/UV lasers. In a near future we anticipate a significant improvement of this concept, e.g. by using waveguiding technique to increase the length of the amplifier and thus boost up the SXRL output energy by at least one order of magnitude (15). Our measurements suggest that thanks to the perfect wavefront it should be possible to focus the 32.8 nm seeded laser into a near diffraction-limited spot, and thus achieve a soft x-ray intensity close to 1015 W.cm<sup>2</sup>. The superb spatial beam quality and full longitudinal coherence make this source an excellent scientific tool for applications such as soft x-ray holography, phase contrast imaging, and microscopy.

## References

1. Tesla FEL reports, [http://flash.desy.de/reports\\_publications/index\\_eng.html](http://flash.desy.de/reports_publications/index_eng.html) /
2. J. Seres et al., Nature 433 , 596 (2005)
3. X. Zhang et al. , Opt. Lett. 29, 1357 (2004)
4. E. Takahashi et al., Phys. Rev. A 66, 021802 (2002)
5. J.-F. Hergott et al., Phys. Rev. A 66, 021801 (2002)
6. H. Daido, Rep. Prog. Phys. 65, 1513 (2002)
7. D.L. Matthews et al., Phys. Rev. Lett., 54, 110 (1985).
8. P.V. Nickles et al., Phys. Rev. Lett. 78, 2748 (1997)
9. S. Sebban et al., Phys. Rev. Lett. 86, 3004 (2001).
10. J.J. Rocca et al., Phys. Rev. Lett. 73, 2192 (1994)
11. T. Ditmire et al., Phys. Rev. A 51, 0R4337 (1995)
12. Ph. Zeitoun et al., Nature 431, 466 (2004)
13. Y. Wang et al., Phys. Rev. Lett. 97, 123901 (2006)
14. S. Sebban et al., Phys. Rev. Lett. 89 253901-1 (2002)
15. B. Cros et al., Phys. Rev. A 73, 033801 (2006)

16. J. Ph. Goddet et al., *Opt. Lett.* 32, 1498 (2007)
17. T. Mocek et al., *Phys. Rev. Lett.* 95, 173902 (2005)
18. S. Sebban et al., *JOSA B* 80, 195 (2003)
19. O. Guilbaud et al., *Europhys. Lett.* 74, 823 (5)
20. W.H. Southwell, *J. Opt. Soc. Am.* 70, 998 (1980)
21. S. Le Pape et al., *Phys. Rev. Lett.* 88, 183901-1 (2002)
22. A. Marechal, *Rev. d'Optique*, 26, 257 (1947)
23. G. Maynard et al., *Contrib. Plasma Phys.* 47, No. 4–5, 352 (2007)
24. B. Talin et al., *Phys. Rev. A* 51, 1918 (1995)
25. J.A. Koch et al., *Phys. Rev. A.* 50, 1877 (1994)