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Table-top femtosecond soft X-ray laser by collisional ionization gating

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The advent of X-ray free-electron lasers has granted researchers an unprecedented access to the ultrafast dynamics of matter in the nanometer scale^{1, 2, 3}. Aside from being compact, seeded plasma-based soft X-ray lasers (SXRL) turn out enticing as photon-rich⁴ sources (up to 10^{15} per pulse) displaying high-quality optical properties^{5, 6}. Hitherto, the duration of these sources has been limited to the picosecond range⁷ consequently restricting the field of applications. This bottleneck was overcome by gating the gain through ultrafast collisional ionization in high-density plasma generated by an ultra-intense infrared pulse (a few 10^{18} W.cm⁻²) guided in an optically pre-formed plasma waveguide. For electron densities ranging from 3×10^{18} cm⁻³ to 1.2×10^{20} cm⁻³, the gain duration was measured to drop from 7 ps to an unprecedented value of about 450 fs, paving the way to compact and ultrafast SXRL beams with performances previously only accessible in large-scale facilities.

In plasma-based soft X-ray laser (SXRL) systems, the amplifying medium is a hot, highly charged plasma generated by the interaction of an intense laser pulse with a solid⁸ or a gas⁹ target. Hitherto, in all SXRL amplifiers operating at saturation, population inversion between the levels of the lasing ion is induced by electron collisional excitation, resulting in strong amplification of spontaneous emission (ASE) down to 3.56 nm¹⁰ with high temporal coherence¹¹. When seeded with a high order harmonic (HH) beam, the generated soft X-ray beam exhibits excellent spatial properties⁶ and adjustable linear polarization¹². However, all previous attempts have failed in shortening the duration of plasma-based SXRL below one picosecond^{13, 14}, the fundamental lower limit being governed by the very narrow gain bandwidth of collisionally pumped plasma amplifiers. Therefore, a wealth of phenomena occurring on femtosecond timescales, such as molecular dynamics² or lattice vibrations³, remained unfortunately out of reach for these attractive sources.

To overcome this constraint, recent numerical calculations¹⁵ proposed a transposition of chirped pulse amplification technique to the soft X-ray range. By stretching a femtosecond HH seed to duration close to the gain lifetime, this would allow continuously and coherently extracting the energy stored in the plasma. After compressing the amplified seed, this scheme theoretically inferred SXRL pulses as short as 200 fs but has not been experimentally demonstrated so far. Other research directions have been considered to bring SXRL in the ultrafast domain, mainly based on straightforward approach capitalizing on intrinsically femtosecond population inversion schemes. The so-called recombination scheme in plasmas is an attractive candidate¹⁶, nevertheless it requires drastic plasma conditions, which have not been implemented to date. More recently, the inner-shell ionization scheme has demonstrated an ultrafast capability in the keV range, however this scheme requires a very intense hard X-ray pump to work efficiently^{17, 18}.

Our approach is based on the collisional ionization gating (CIG) of the gain media to reduce the time window in which the lasing action takes place. In the present work, an ultrashort infrared laser pulse (5×10^{18} W/cm²) was focused onto a krypton gas jet to generate the lasing ion species through optical-field ionization¹⁹ (OFI). The resulting electron distribution permits to achieve a population inversion by collisional pumping leading to the lasing of the $3d^9 4d_{j=0} \rightarrow 3d^9 4p_{j=1}$ transition of nickel-like krypton at 32.8 nm¹⁰. In this scheme, the amplifier lifetime

strongly depends on the depletion rate of the lasing ion population²⁰ due to collisional ionization during the lasing process.

When increasing the plasma density, this process becomes fast enough to provoke an anticipated interruption of amplification in less than a few hundred femtoseconds, consequently shortening the duration of the soft XRL pulse. As shown in numerical calculations from Figure 1, increasing the electron density from $6 \times 10^{18} \text{ cm}^{-3}$ up to $4 \times 10^{20} \text{ cm}^{-3}$, CIG process becomes sufficiently fast to exhaust the gain in less than 100 fs, thus opening a promising path toward ultrafast soft X-ray lasers. Furthermore, increasing the density leads to a dramatic boost of the lasing yield caused by the enhancement of the gain coefficient and the saturation intensity (see S.I.), pushing previous limitations in terms of peak brightness of compact SXRL sources⁷.

The propagation of the driving laser in near-critical density plasma over several millimetres is the most serious challenge to generate a saturated plasma amplifier. In such a dense OFI plasma, diffraction, refraction and self-focusing of the pump beam limit the amplifying length to a few hundreds of micrometers²¹. Our approach overtakes those difficulties by implementing an optically preformed plasma waveguide^{22, 23}. It is created by a sequence of IR short ("ignitor", 130mJ, 30 fs) and long ("heater", 690 mJ, 600 ps) laser pulses focused into a high-density gas jet by an axicon lens (Figure 2). The ignitor-heater sequence permits to first pre-ionize the gas, generating free electrons that are later heated by the second pulse and then multiplied through cascade collisional ionization. The following plasma hydrodynamic expansion produces a density channel that is appropriate for guiding the high intensity driving laser pulse. Advantageous guiding conditions allowed strong amplified spontaneous emission from the plasma (see S.I.).

To evaluate the influence of electron density on the temporal properties of the 32.8 nm lasing emission, we have measured the temporal gain dynamic by seeding the SXRL amplifier using the 25th harmonic of the infrared driving laser. The seed was obtained by focusing a 20 mJ, 30 fs, 813 nm-wavelength laser in a gas cell filled with argon and was image-relayed onto the entrance of the SXRL amplifier using a grazing-incidence toroidal mirror. Strong amplification of the seed pulse was observed when the injection time was synchronized with the gain period. For the highest operated electron density at $1.2 \times 10^{20} \text{ cm}^{-3}$, Figure 3 depicts the far-field beam profile of the seeded SXRL in comparison to the HH signal. Both beams exhibit a nearly Gaussian shape with a FWHM divergence of about 1 mrad. The amplification factor for SXRL is 75 considering

all HH orders and 1000 for only the wavelength-matching HH order (see S.I.). The amplification of the HH portion matching the laser transition at 32.8 nm corresponds to more than three orders of magnitude and the resulting SXRL signal yields about 2 μJ (over 3×10^{11} photons) per shot. Propitious guiding conditions allowed us to enhance the SXRL yield by one order of magnitude over an electron density range from a few 10^{18} to over 10^{20} cm^{-3} , in accordance with our simulations reporting a monotonous increase in gain and saturation intensity (see S.I.).

By varying the time delay between the amplifier creation and the seed pulse injection, we measured the temporal evolution of the amplification factor, thus revealing the actual temporal profiles of the SXRL gain. As shown in Figure 4, the gain duration monotonically decreases from 7 ps to an unprecedented⁸ shortness of 450 fs full width at half-maximum (FWHM) while the amplification peak rises from 150 to 1200, by increasing the plasma densities from $3 \times 10^{18} \text{ cm}^{-3}$ up to $1.2 \times 10^{20} \text{ cm}^{-3}$ respectively. These measurements clearly illustrate the dramatic temporal quenching of the amplification as a result of an increase in electron density. As anticipated from Figure 1, the fast rise of the amplification is due to collisional excitation of the lasing transition taking place after the abrupt field ionization process. The ultrafast decay in amplification brings testimony of the CIG process at high densities. Although vital for pumping, the on-going collisional ionization also mainly contributes to shorten the gain lifetime, the number of lasing ions becoming rapidly scarce due to strong over-ionization.

Using a time-dependent Maxwell-Bloch code²⁴, we undertook modelling of the radiation transfer in the amplifying SXRL plasma relevant to experimental conditions. As shown in Figure 4, our calculations reveal that the amplification life time is shorter and starts earlier with steeper slope for higher gas densities, which is in good agreement with our experimental measurements. Our Maxwell-Bloch numerical model describes well the atomic processes underpinning the gain temporal quenching over a remarkably large range of electron densities covering nearly two orders of magnitude. The final pulse duration of HH-seeded SXRL is Fourier-limited⁵ and therefore shorter than amplification duration (see S.I.). It is thereby determined by the soft X-ray laser transition broadening dominated by electron-ion collisional broadening, particularly strong at the highest densities. The benchmarking of the electron density values used in our numerical model through amplification dynamic as well as interferometry measurements brings high degree of confidence in extracting SXRL final duration from the numerical model. As shown in Figure 5, the temporal structure of the pulse is composed of several periods of Rabi oscillations, induced

by the strong amplification of the pulse. Since the plasma is inhomogeneous (see S.I.), the amplification profile will be a superimposition of different temporal amplification timescales, each corresponding to a particular density. Different parts of the beam will induce Rabi oscillations with slight different periods and only the pulse envelope will be observed²⁵. From our Maxwell-Bloch modelling, the inferred SXRL pulse duration (RMS) varies from 6.4 ± 0.3 ps for $n_e = 3 \times 10^{18} \text{ cm}^{-3}$ down to 123 ± 40 fs (64 ± 21 fs FWHM) for $n_e = 1.2 \times 10^{20} \text{ cm}^{-3}$, thus breaking the decade-long picosecond barrier of plasma-based SXRL.

The achieved combination of shorter duration and boost in photon yield demonstrates more than two orders of magnitude upsurge of plasma-based SXRL output power.

Therefore, the perspective of guiding over longer distances and at higher densities fosters promising opportunities to further outdistance current state-of-the-art performances. Whilst future adequate plasma engineering appears necessary, our calculations show that, despite density inhomogeneities and transverse gradients, SXRL duration as short as 20 fs is expected for $n_e = 4 \times 10^{20} \text{ cm}^{-3}$ (see Figure 5). Combining remarkable beam quality⁶, the full longitudinal coherence⁵ and the jitter-free nature of seeded plasma-based SXRLs, the CIG approach contributes to substantiate the full-fledged capabilities of this compact source to carry out, at the laboratory scale, single-shot high-resolution dynamical studies on the femtosecond time scales^{27, 28}.

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Authors contributions. S. S. proposed the experiment. S. S., J. G, F. T. and A. D. have designed and built the setup. G.M. developed the code OFI-0D. E. O., G. M. and A. L. performed numerical simulations.

J.P.G. and A.T. designed, built and operated the upgraded laser system of “Salle Jaune”. K.T.P., C. T. and P. R. provided support for the operation of facilities. A.F. developed a phase reconstruction and Abel inversion software for the electron density diagnostic.

All authors contributed to analyse data and write the paper. P. Z., S. J., V. M. and A. R. supported the project.

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Figure 1. Computed temporal dependence of the average charge state and gain coefficient of a krypton plasma amplifier for $n_e = 6 \times 10^{18} \text{ cm}^{-3}$ (green), $n_e = 1.2 \times 10^{20} \text{ cm}^{-3}$ (blue) and $n_e = 4 \times 10^{20} \text{ cm}^{-3}$ (red). The grey-tinted part corresponds to the OFI regime preceding the subsequent collisional ionization. The dotted lines report the increase of the ionization rate with density. Therefore, at high densities, the steep depletion in the number of Kr^{8+} ions leads to an ultrafast gating of the gain lifetime (CIG process). The yellow area illustrates the region where Kr^{8+} ions are predominant.

Figure 2. Schematic of the experimental arrangement. The waveguiding beam is composed of a sequence of a short (130 mJ, 30 fs) and a long pulse (690 mJ, 600 ps). It is being focused over the whole jet length thanks to an axiconic lens and creates, following collisional ionization and hydrodynamic expansion, a plasma channel. Then, the pump beam (1.36 J, 30 fs) is being focused at the entrance of the channel with a spherical mirror and being guided afterwards. Hence, an amplifier with Kr^{8+} lasing ion species over the whole gas jet length is implemented. A third IR beam (16 mJ, 350 fs) is used to generate high-harmonics seed in an argon-filled cell. The latter are image-relayed onto the entrance of the plasma and synchronized with the gain lifetime.

Figure 3. Far-field beam profiles of the HH (a) and the seeded SXRL (b) signals in case of $n_e = 1.2 \times 10^{20} \text{ cm}^{-3}$. The SXRL energy is assessed at $2 \mu\text{J}$, which corresponds to more than 3×10^{11} coherent photons. The 1 ± 0.2 mrad divergence and nearly Gaussian beam profile of HH is maintained over the plasma amplification. A factor of about 75 is reported between total HH and seeded SXRL signals.

Figure 4. Temporal dependence of the amplification factor with respect to the seeding delay. Experimental (blue circles) and Maxwell-Bloch modelling results (red squares) for a scan in plasma density: $n_e = 3 \times 10^{18} \text{ cm}^{-3}$, $n_e = 7.9 \times 10^{18} \text{ cm}^{-3}$, $n_e = 3.2 \times 10^{19} \text{ cm}^{-3}$ and $n_e = 1.2 \times 10^{20} \text{ cm}^{-3}$. Because of the inhomogeneity of the plasma channel, lower density regions, for which the gain duration is longer, are accountable for the experimental curve end tail.

Figure 5. Simulated temporal profiles of the amplified SXRL for the set of studied densities and a prospective one of $n_e = 4 \times 10^{20} \text{ cm}^{-3}$ (red). The duration strongly depends on the density and ranges from $6.4 \pm 0.3 \text{ ps}$ to $50 \text{ fs} \pm 18 \text{ fs RMS}$. The highest experimental density (blue) results in an amplified beam with duration of $64 \pm 21 \text{ fs FWHM}$ ($123 \pm 40 \text{ fs RMS}$). The curves have been normalized to one and their RMS duration specified.

Methods

Laser system and target. The experiment was conducted at Laboratoire d'Optique Appliquée with the “Salle Jaune” Ti:Sapphire laser system, which delivers $5 \text{ J} / 30 \text{ fs}$ (FWHM) pulses at 1 Hz centered around a wavelength of 813 nm . The main laser beam is divided into four beams: a pump beam, a waveguiding beam, a HH driver beam and a probe beam. The waveguiding beam is composed of two pulses: the ignitor (130 mJ , 30 fs FWHM) arriving 600 ps before the heater (690 mJ , 600 ps FWHM). The gas nozzle has a rectangular section of $500 \mu\text{m} \times 5 \text{ mm}$. The backing pressure of krypton gas jet has been set at 150 bar whereas the valve is opened 10 ms before the main pulse over a period of 8 ms .

At the highest reported density, appropriate conditions for guiding in terms of size and transverse density gradient were granted for the pump beam focused about 1.55 ns after the arrival of the “ignitor”. For investigation at lower densities, for which waveguiding is not needed, a 5 mm -long krypton-filled gas cell was used.

Plasma amplifier. The amplifying medium is achieved focusing the pump beam pump beam using a 75 cm -focal length spherical mirror into the krypton gas jet. The beam was turned circularly polarized thanks to a quarter-wave plate. This allows catering for hot electrons needed to efficiently pump the laser transition. The measured focal spot was about $38 \mu\text{m}$ in $1/e^2$ -diameter, corresponding to an intensity on target of about $5 \times 10^{18} \text{ W/cm}^2$.

The soft X-ray seed is implemented focusing the HH driver beam into a gas cell of variable pressure and length, filled with argon. The linearly polarized beam was focused using a 70 cm -focal length plano-convex lens. The intensity upon target is assessed at about 10^{15} W/cm^2 . The

25th harmonic signal was then spectrally tuned onto the amplifier amplified spontaneous emission (ASE) laser line at 32.8 nm chirping the beam HH driver beam while its flux was optimized changing the gas pressure and altering the focusing conditions, notably by fine-tuning the beam aperture with a motorized iris. The appropriate conditions were found for a 12-mm long cell with a pressure of 40 mbar and a focal plane situated about 5 mm after the cell entrance. The seeding was fulfilled re-imaging the harmonic output onto the entrance of the amplifier plasma, using a 5 degrees grazing incidence gold-coated toroidal mirror in a 1.5 m - 1 m conjugation. The experiment was aligned making the HH beam collinear with the infrared pump and waveguiding beams. The spatial coupling of the HH beam and the SXRL gain region was done in the IR superposing the HH driver with the focal spot of pump. The temporal overlap between the HH signal and the SXRL amplification period was approached synchronizing the HH driver and the pump beams thanks to a 30 fs-accuracy delay line.

The waveguide transmission was assessed between 40 and 50% integrating the whole energy of the beam profile imaged at the entrance and the output of the plasma channel.

X-ray beam diagnostics. The XUV spectrometer consists of a removable on-axis deflecting soft X-ray plane mirror, a gold-coated 400 mm-focal length spherical mirror at 77.5 and 80 degrees incidence respectively, along with a 5 mm × 5 mm grating with 1000 lines/mm and a soft X-ray 16 bit charge-coupled device (CCD) camera of 2048 × 2048 pixels resolution. The energy distribution measurement was performed using a 25 degrees off-axis parabola of focal length 200 mm, a deflecting plane mirror at 80 degrees incidence and a soft X-ray camera of 1024 × 1024 pixels resolution. The interferential multilayer coating of the parabola designed to yield high reflectivity at 32.8 nm was deposited at Institut d'Optique, France. Two aluminium filters of 2 × 0.15 μm and 0.1 μm have been used to isolate XUV from IR radiation. The amplification factor, A, has been computed integrating the energy distribution of the signals (see S.I.) using the formula:

$$A = (N_{XRL} - N_{ASE} - N_{HH}) / N_{HH}$$

where N_{XRL} , N_{ASE} and N_{HH} are the number of counts of the seeded XRL, ASE and HH signals respectively, averaged over 3 or 4 shots.

Electron density mapping. A Mach-Zehnder interferometer makes a side-view electron density

probing the plasma doubling the laser frequency with a beta barium borate crystal to limit the effect of refraction within the medium and increase the highest measurable electron density. The interferograms were recorded using a 16-bit CCD camera with a 13.3-magnification optical setup imaging a plasma zone of 700 μm long by 500 μm at the entrance of the waveguide.

Numerical modelling. The field and collisional ionization of the krypton plasma has been characterized in Figure 1 using OFI-0D code, which has been derived from COFIXE code²⁹ taking account of 92 atomic levels of krypton. It allows modelling the plasma amplifier in one point in the space notably in terms of ion populations, gain, emissivity and saturation intensity with respect to key parameters, such as gas pressure, laser intensity and field polarization. Macroscopic phenomena like hydrodynamic expansion along with radiative losses and transfer are not considered here. The calculations were performed for a krypton plasma amplifier generated by a circularly polarized laser beam with a peak intensity of $5 \times 10^{18} \text{ W/cm}^2$.

The propagation and amplification of the high harmonic beam inside the plasma amplifier was modelled with our time-dependent Maxwell-Bloch model DeepOne³⁰. The evolution of plasma parameters and atomic level populations and rates were computed using an advanced atomic physics model. This data (temporal evolution of the electron density and temperature, atomic collisional radiative rates, population of the lasing ion and population of several atomic levels strongly coupled to the lasing levels) was fed to our time-dependent Maxwell-Bloch code so as to obtain with its simplified three-level atomic model the same population inversion as the more complete atomic physics package. The Amplified Spontaneous Emission was taken into account consistently via a stochastic source term. Due to the induced stochasticity, each point in Figure 5 was averaged over 50 simulations. Because of uncertainty on the energy of the seed beam, the amplification given by our model was normalized to the experimental data using the maximum amplification point. The duration of the amplified pulse is taken to be the full width at half maximum of the pulse envelope (as shown in Figure 5). In addition to this, due to the complex temporal profile of the pulse, the root-mean-square duration was also computed.







