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Observation of spectral gain narrowing in a high-order harmonic seeded soft-x-ray amplifier

F. Tissandier,* S. Sebban, M. Ribière, J. Gautier, Ph. Zeitoun, G. Lambert, A. Barszczak Sardinha, J.-Ph. Goddet, F. Burgy, T. Lefrou, C. Valentin, and A. Rousse

Laboratoire d'Optique Appliquée, ENSTA Paristech/Ecole Polytechnique/CNRS UMR 7639, F-91761 Palaiseau Cedex, France

O. Guilbaud and A. Klisnick

Laboratoire d'Interaction du Rayonnement X Avec la Matière, Université Paris Sud/CNRS UMR 8624, F-91495 Orsay Cedex, France

J. Nejdl and T. Mocek

Department of X-ray Lasers, Institute of Physics, Na Slovance 2, CZ-182 21 Prague 8, Czech Republic

G. Maynard

Laboratoire de Physique des Gaz et des Plasmas, Université Paris Sud/CNRS UMR 8578, F-91405 Orsay Cedex, France

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We report an observation of spectral gain narrowing of a high-order harmonic amplified by a soft-x-ray optical-field-ionized plasma. The temporal coherence and spectral linewidth of both the seeded and unseeded soft-x-ray lasers were experimentally measured using a varying-path-difference interferometer. The results showed that the high-order harmonic is subject to a strong spectral narrowing during its propagation in the plasma amplifier without rebroadening at saturation. This is in good agreement with a radiative transfer calculation including gain narrowing and saturation rebroadening.

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Table-top high-repetition-rate soft-x-ray lasers (SXRLs) are promising short-wavelength sources for emerging applications such as lithography, high-resolution microscopy, or holography. While common soft-x-ray lasers working in the amplification of spontaneous emission (ASE) regime usually present low-quality spatial profiles and spatially incoherent beams, these same laser amplifiers seeded by a fully coherent high-order harmonic of an infrared laser offer many more perspectives in terms of beam quality and coherence [1,2]. When the amplifier is properly seeded, the spatial quality of the output beam is dramatically enhanced. It has been demonstrated to be highly collimated, with a Gaussian transverse profile, and spatially coherent [1,3,4], up to the diffraction limit [5,7]. In addition to being more energetic than either the ASE or the unamplified harmonic, a seeded soft-x-ray laser pulse is temporally fully coherent, thus having a duration (a few picoseconds [6]) predominantly limited by Fourier transform [8]. In this work, the temporal coherence and output energy of a seeded soft-x-ray laser emitting at 32.8 nm have been measured for different amplifier lengths. The results have been compared to numerical simulations based on a simple collisional-radiative model.

The optical-field-ionized (OFI) soft-x-ray amplifier under investigation was generated in a configuration similar to that described in [1]. The pump laser is an infrared (ir) multiterawatt Ti:sapphire system providing two independent 35-fs pulses at a central wavelength of 815 nm with a maximum repetition rate of 10 Hz. The soft-x-ray amplifier is generated by focusing the circularly polarized beam delivering around 600 mJ into a gaseous low-density krypton cell with a 1-m-focal-length spherical mirror. Collisional excitation is due to the hot free electrons of the Ni-like krypton plasma and induces strong laser gain on the transition at $\lambda_0 = 32.8$ nm [9]. This amplifier medium generates a soft-x-ray laser in the ASE regime. It is seeded by a high-order harmonic (HOH) beam generated by the second ir beam. This beam, consisting of linearly polarized 10-mJ pulses, is focused by a 1.5-m-focal-length lens into a gas cell filled with 30 mbar of argon. The HOH beam is coupled into the amplifier by a grazing-incidence toroidal mirror imaging the source with a magnification of 1.5. In the ASE regime, the SXRL beam has a strong divergence (15 mrad) and exhibits a speckled far-field profile. When properly seeded by the HOH beam, the SXRL beam is well collimated (divergence of 1 mrad), with an Airy far-field pattern. The most effective configuration in terms of SXRL energy was found for a krypton pressure of 30 mbar, a plasma length of 6 mm, and a delay between the creation of the amplifier and the injection of the HOH pulse of 3 ps. Unless otherwise noted, the results presented in this paper were all obtained with this set of parameters.

We have investigated the spectral bandwidth of the SXRL. Because of its extremely narrow linewidth, the spectral profile cannot be resolved by the transmission grating spectrometer. We used instead an interferometric method based on the measurement of the temporal coherence of the SXRL pulse. The SXRL beam is directed toward a variable path difference interferometer based on the Fresnel mirror system. Detailed information on this soft-x-ray interferometer can be found in [10]. The interferometer produces fringes that are recorded on an extreme uv charge-coupled device (CCD) camera. A path difference between the two interfering beamlets can be introduced without laterally moving them, ensuring that any loss in fringe visibility is not due to a loss of spatial coherence. Its settings were such that the maximum time difference we could introduce was around 6.5 ps, allowing the spectral profile

*fabien.tissandier@ensta.fr

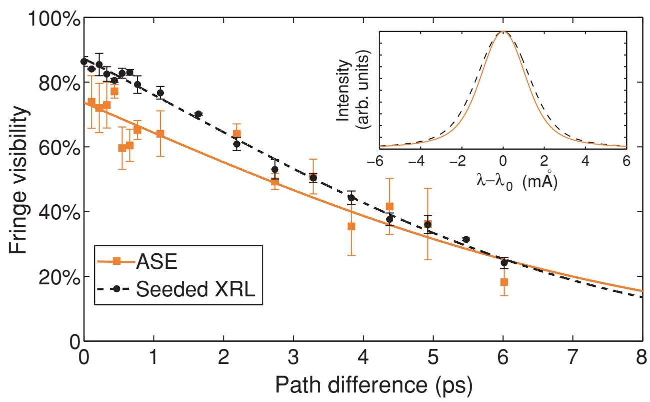

FIG. 1. (Color online) Visibility of the interference fringes as a function of the time difference for the ASE and seeded SXRL. The lines represent the result of fitting by a relevant function (defined in text). Inset: Reconstructed spectra from Fourier transforms of the fit functions.

of the radiation to be reconstructed by Fourier transform with a resolution power better than 2×10^5.

The experimental measurement of fringe visibility variations with the optical path difference is presented for both the ASE and the seeded SXRL in Fig. 1. Each point results from an average over around ten shots and the error bar indicates the standard deviation. Note that the fringe visibility is lower than 100%. This is not due to a degradation of the harmonic beam spatial coherence by the plasma [3] but rather to the experimental setup. A fringe actually covers around 5 CCD pixels, causing a loss of information. The presence of remaining (spatially incoherent) ASE may further deteriorate the visibility. To ensure that the laser line spectral profile is the most general, that is, a Voigt profile, the experimental data were accurately fitted by an analytical function defined as the product of a Gaussian function and a decreasing exponential function (dotted lines). The coherence time τ_c of the pulses, defined by the time difference at which the visibility is decreased by a factor $1/e$, is not the same for the two cases investigated. It was estimated as 5.1 ± 0.2 ps for the seeded SXRL pulse and slightly larger at 5.5 ± 0.3 ps for the ASE SXRL pulse. According to the Wiener-Khinchin theorem, the evolution of the fringe visibility with the time difference is the Fourier transform of the spectral density of the source. The spectral profiles have been calculated from the fitted visibility evolutions, and correspond to spectral Voigt profiles with a full width at half maximum (FWHM) of $\Delta\nu = 89 \pm 6$ GHz ($\Delta\lambda = 3.2 \pm 0.2$ mÅ) for the seeded SXRL and $\Delta\nu = 75 \pm 8$ GHz ($\Delta\lambda = 2.7 \pm 0.3$ mÅ) for the ASE. Seeding with a harmonic produces a pulse with an initial bandwidth much larger than the ASE bandwidth and more of that bandwidth is preserved. While the difference in bandwidth stays within the error bars, this result was numerically confirmed. This issue will be discussed later in the paper.

The spectral width of the SXRL line before amplification (optically thin) is determined by homogeneous (natural) and inhomogeneous broadening. The processes responsible for homogeneous broadening include radiative decay (natural broadening) and electron collision-induced transitions. They lead to a Lorentzian line profile calculated with a collisional-radiative model taking into account the non-Maxwellian electron energy distribution [11]. Its FWHM is $\Delta\lambda_H = 5$ mÅ in the case of the 32.8-nm Ni-like laser. The Stark effect has been introduced as a source of inhomogeneous spectral broadening, but numerical simulations have shown that it can be neglected [12]. The main process responsible for inhomogeneous broadening is the ion Doppler effect. Ions are indeed rapidly heated after ionization by the laser field. Theoretical investigations [12] have shown that strong correlations exist in OFI plasmas, which rapidly relax into an uncorrelated form with a characteristic time equal to the plasma period (a few hundreds of femtoseconds). The ion equilibrium temperature in the case of our low-density OFI plasma is $T_i = 6$ eV. This leads to an inhomogeneous Gaussian line profile with a FWHM of $\Delta\lambda_D = 7$ mÅ for the 32.8-nm Ni-like laser. These very low homogeneous and inhomogeneous broadening values along with radiative transfer explain the strong monochromaticity of this type of OFI-pumped SXRL ($\Delta\lambda/\lambda \simeq 10^{-5}$).

Using a variable-length gas cell, we have been able to measure the temporal coherence of the seeded SXRL for plasma lengths of 1, 2, 4, and 6 mm. For lengths shorter than 6 mm, the measurements were not possible in the ASE regime because of a weak signal level. The calculated spectral density from the fitted visibility evolutions is plotted in Fig. 2 for each plasma length. The unamplified harmonic spectrum is not represented here. We were nevertheless able to measure it using the soft-x-ray spectrometer. Its FWHM was evaluated to be $\Delta\lambda_{HHG} = 1.5 \pm 0.3$ Å, which is much larger than after even only 1 mm of amplification ($\Delta\lambda = 4.8 \pm 0.5$ mÅ). Because of the very small bandwidth of the SXRL line, the harmonic is subject to a strong spectral narrowing which—as a consequence of the very high gain value (60 cm^{-1})—takes place after a very short distance of propagation. It can be deduced that much of that spectral narrowing occurs during the first hundreds of micrometers of propagation or even less, since from 1 mm of propagation to 6 mm the spectral linewidth has the same order of magnitude. Note that the gain saturation is reached after 2–3 mm of propagation, as will be shown later. We did not observe any saturation rebroadening, which suggests that the homogeneous broadening contribution is predominant at saturation [13]. The Voigt line profile also

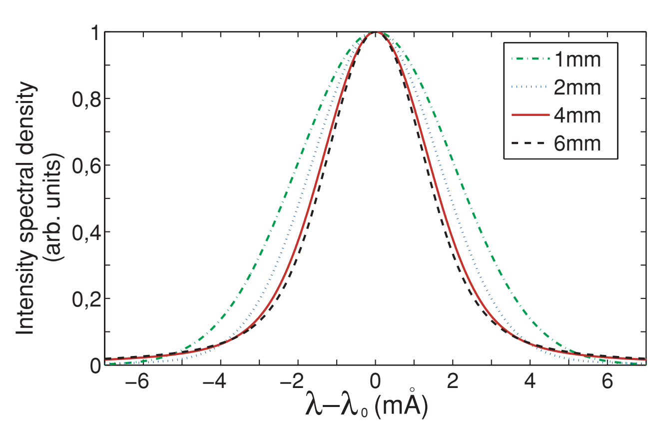

FIG. 2. (Color online) Spectral density of the SXRL laser line at different propagation lengths in the plasma.

seems to tend more to a Lorentzian profile for the longest propagation lengths in the plasma.

We used an existing numerical code based on radiative transfer where gain narrowing and possible saturation rebroadening are taken into account to study this spectral evolution [8,13]. This model does not account for the coherent interaction of the high-order harmonic with the plasma population inversion: When the amplifier has a much shorter bandwidth than the seed, Rabi oscillations of the population are expected to appear at saturation and lead to electric field temporal modulation and thus to sidebands in the spectral profile. No strong evidence of oscillations has been found in our experimental data. Starting from some theoretical predictions [14,15], we nevertheless calculated the expected fringe visibility evolution with the path difference in the presence of this phenomenon and found that the visibility modulation would be of the order of the experimental fluctuations. The presence of inhomogeneous broadening may also attenuate the amplitude of the oscillation. In our model, the optically thin profile of the SXRL line was a Voigt convolution of the respective inhomogeneous and inhomogeneous contributions as given above. The spontaneous emission was supposed to have the same profile as the small-signal gain. The initial HOH spectrum was assumed to be Gaussian with a width equal to the experimental value. The gain was set to 60 cm^{-1}, and the intensity of the harmonic was chosen to obtain amplification values close to experimental ones: For a length of 6 mm the amplification factor (seeded SXRL intensity corrected for the ASE and divided by the harmonic intensity) is 200. The results of this study are presented Fig. 3.

First, the evolution of the seeded SXRL intensity with the amplification length is plotted (right *y* axis), calculated with the same code (dashed line) as well as experimentally measured (triangle markers). The intensity was measured using the soft-x-ray spectrometer by integration over the harmonic—or amplified harmonic—line. Each data point corresponds to an average over several shots, and the error bar indicates the standard deviation. No amplification is visible for a length

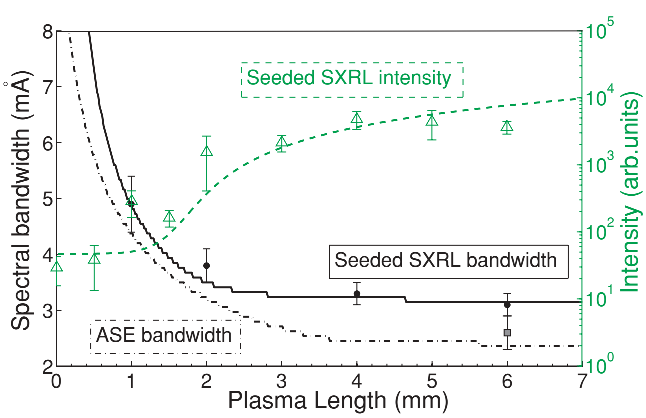

FIG. 3. (Color online) Spectral bandwidth of the seeded SXRL and the ASE (left *y* axis) and intensity of the seeded SXRL (right *y* axis) as a function of the propagation length in the plasma amplifier. The markers stand for experimental data and the lines result from numerical simulations.

$\lesssim$1 mm because the intensity is integrated over a spectral bandwidth larger than the transition linewidth. This length is actually the length over which gain matching occurs, so that only a very small spectral slice of the harmonic is amplified, leading to an overall amplification negligible compared to the total intensity of the harmonic. We can also see that saturation is indeed very shortly reached, after 2–3 mm of amplification. For longer cell lengths, some reabsorption by residual neutral or weakly ionized krypton may occur, explaining why the experimental signal does not further increase with length. Corresponding to the left *y* axis is plotted the calculated evolution of the spectral bandwidth for the ASE (dash-dotted line) and the seeded SXRL (solid line) along with the experimental data (markers). It can be seen that the agreement between measurements and calculations is very good. The simulation confirms that the harmonic is very rapidly spectrally narrowed over the gain-matching length and that, when saturation is reached, its bandwidth decreases very slowly with increasing amplification length. It also confirms that there is no rebroadening at saturation for either the ASE or the seeded SXRL.

We have also numerically investigated the dependence of the SXRL spectral bandwidth on the harmonic injection level. As expected, the bandwidth increases with the harmonic fluence. Seeding with a HOH allows more energy to be extracted by use of the whole population of the lasing ions, thus slightly increasing the spectral bandwidth. These bandwidth variations are very limited. In our experiment, the harmonic fluence was estimated to be around 600 nJ/cm^2. It was calculated that an increase of three orders of magnitude of that fluence, corresponding to a strong microjoule harmonic, would lead to a SXRL bandwidth of 5 mÅ. Since the pulse reached the Fourier limit [8], that would also lead to a slight decrease of the pulse duration. These very small variations indicate nonetheless that the seeding of a soft-x-ray amplifier with a strong harmonic is not sufficient to effectively reduce the pulse duration below the picosecond range. Other approaches, such as overcoming spectral narrowing by increasing the Doppler broadening, are required.

In conclusion we report in this paper a demonstration of gain-induced spectral narrowing in a high-order harmonic seeded soft-x-ray laser. By measuring the spectral profile of this 32.8-nm Ni-like krypton OFI laser at different plasma lengths, we were able to study the variations of the spectral linewidth along the propagation in the amplifier, showing a rapid narrowing over a very short length followed by a slight linewidth decrease at saturation, without rebroadening. We were also able to compare the saturated linewidths in the seeded and ASE regimes. In addition, the data have been supported with numerical simulations including a non-negligible Doppler broadening owing to the rapid ion heating of the initially correlated OFI plasma. The calculated linewidth evolutions were in good agreement with our experimental data.

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