AN INTENSE KHz TWO-COLOUR HIGH HARMONIC SOURCE FOR SEEDING FELS FROM EUV TO SOFT X-RAY RANGE

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Abstract

Free-electron lasers have been recently evolving very fast in the extreme-ultraviolet to soft X-ray region. Once seeded with high harmonics, these schemes are considered as next generation soft X-ray light sources delivering ultrashort pulses with high temporal and spatial coherence. Here we present a detailed experimental study of a kHz two-colour (fundamental + second harmonic) high harmonic generation and investigate its potential as a suitable evolution of the actual seeding sources. It turns out that this source (both odd and even harmonics) is highly tuneable, and delivers intense radiations with only one order of magnitude difference in the photon yield from 65 nm to 13 nm. Then, the implementation of this technique on seeded FELs would allow amplifications to be achieved at wavelengths shorter than previously accessible.

INTRODUCTION

Seeding Free-Electron Lasers (FEL) [1]-[2] with High order Harmonics (HH) generated in gas from the VUV (Vacuum Ultra Violet, 80-130 nm) to the soft X-ray (10-40 nm) region is a quite recent but emerging topic. Indeed, it has already opened significant perspectives for developing the future ultimate relatively compact coherent jitter-free and aberration-free source in order to observe the ultrafast dynamics of matter at nanometre scale. Seeding relies on benefits from the high-quality properties of the HH source, particularly the ultrashort pulse duration or the high degree of spatial and temporal coherence.

Since the beginning of this century, novel linear accelerator-based single-pass FEL sources have been emerged around the world. Presently, different sites have implemented or plan to implement soon seeding schemes in their facilities as a main mode of functioning. The first proof-of-principle seeding experiment has been done in 2007 at 160 nm [1] at the SCSS (Japan) Test Accelerator [3]. The next scheduled test-bed HH seeding facility is SPARC (Italy), which is almost ready to deliver photons in the VUV region down to 114 nm. sFLASH (Germany) is also now being designed and should operate in 2010 in the soft-X ray region from 30 nm to 13 nm. FERMI (Italy) and MAX-IV (Sweden) FELs also expect to use 20 nm-30 nm HH sources for seeding. Other projects, such as SPARX (1.5 nm, Italy), ARC-EN-CIEL (0.8 nm, France) and PSI-XFEL (0.1 nm, Switzerland) consider seeding as a valuable option for X-rays. In these proposals, some additional amplification stages of FEL harmonics of the seed, like in high-gain harmonic-generation (HGHG) [4] and cascade configurations [5] will be coupled to the soft X-ray seed injection.

Essentially, the seeding configuration in FEL is a direct evolution of the classical Self Amplified Spontaneous Emission (SASE) [6], which provides a very high brightness emission (maximum of 100 μJ in pulses of less than 100 fs duration) at short wavelengths but with a limited temporal coherence. The spectral and temporal profiles are typically composed of a series of randomly distributed spikes, reflecting the stochastic nature of the SASE generation process that is amplification of noise. In seeding arrangements, the amplification is triggered by the strong harmonic seed rather than the noise floor leading to strong and coherent amplified FEL radiation.

The proof-of-principle experiment showed that the 160 nm light could be coherently amplified, while the seeded energy per pulse was on the order of nanojoule [1]. The single shot seeded emission achieved then three orders of magnitude higher intensity than the unseeded one and a factor of 500 compared to the seed, almost reaching the microjoule level. Moreover, the corresponding spectral distribution presented a regular quasi perfect Gaussian shape.

As most of the applications are aiming in the wavelength range from soft to hard X-ray (<1 nm), valuable HH seed intensity is required in this spectral range, where unluckily the efficiency of the classical harmonic generation, from 800 nm Ti: Sa laser system, has a sharp decrease.

Finally, seeding with high harmonics should not have any constrain on the machine performances, such as the repetition rate and the tuneability. This is why there is an urgent need of developing new HH sources, more intense at shorter wavelengths, with higher repetition rates, larger tuneability, and with an easy control of the injection parameters (wavelength, intensity, pulse duration…).

Harmonics generated in an orthogonally polarised two-colour laser field [7]-[8], consisting of mixing the fundamental frequency (ω, actually the fundamental pulsation) and its second harmonic (2ω), should fulfil all the above-mentioned seeding requirements.

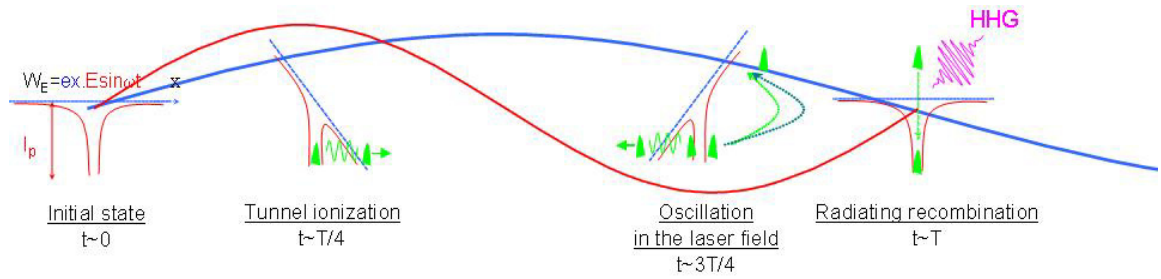

Figure 1: Semiclassical model in three steps, explaining the high order harmonic generation process (HHG). The red line represents the IR driving laser fundamental frequency field and the blue line the field of its second harmonic. T is the optical period of the driving laser.

HIGH HARMONICS GENERATED IN A TWO-COLOUR FIELD

While in classical harmonic generation electrons are extracted, accelerated and radiatingly recombined every half optical cycle of the IR driving laser field (red line, Fig. 1), in a two-colour field (the second harmonic of the fundamental frequency being the bleu line) all the process occurs only once every optical cycle. As a consequence, spectra do no more present only odd harmonics but also even harmonics, corresponding to the odd harmonics naturally generated by 2ω ($2\times(2n+1)$), and to the harmonics coming from the mixing itself ($2\times(2n)$). "n" is an integer.

Second, there is a "redshift" of the whole spectrum, i.e. a shifting to lower harmonic orders of the cut-off, the region where the number of photons start to decrease fast. In a standard harmonic generation process, the spectral energy position of the cut-off, $E_{cut\text{-}off}$ (equation 1), is mainly determined by the maximum of energy gained by the tunnelled electrons, oscillating in the laser field. This energy is equal to $3.2U_P$, where U_P (equation 2) is the ponderomotive potential.

$$E_{Cut-off} = I_P + 3.2U_P \quad (1)$$

$$U_p \propto I_{Laser}\lambda^2_{Laser} \quad (2)$$

Where λ_{Laser} and I_{Laser} are respectively the wavelength and intensity of the laser which generates the harmonics. I_P is the ionisation potentials of atoms.

In our mixing case, a first approach to explain the general behaviour would lead to simply consider that the laser is neither the IR beam nor the blue beam, but a "mixed" beam, which intensity and wavelength are bounded by the ω and 2ω ones. As a consequence, with the presence of the 2ω element, the maximum of energy reached by the electrons is decreased, as U_P is squarely dependent on the wavelength of the laser. This implies an increase of the cut-off position in term of wavelength, which is unwished for a seeding.

Third, the efficiency of generation can be highly increased. The raise has been explained using the semiclassical three-step model; the two-colour laser field induces a selection of the short electron quantum path, for which the specific moment when the electrons are released from the atom corresponds to a 10 times higher ionisation rate than with the single-colour field [9].

EXPERIMENTAL SET-UP

The two-colour (ω+2ω) harmonic generation experiment has been performed at the Laboratoire d'Optique Appliquée (France) by means of a kHz Ti: Sa laser system at 800 nm (ω) delivering maximum of 7 mJ energy in 35 fs FWHM (Full Width Half Maximum) pulses. Fig. 1 presents a scheme of the set-up. In order to generate the 400 nm radiation (2ω), a BBO (Beta Barium Borate, type 1) doubling crystal, is directly inserted in the IR beam path between a 1.5 m focusing lens and a gas cell (typically 4 to 7 mm long). In this geometry, the second harmonic component propagates along the same axis as the IR beam, and consequently the spatial overlap between the ω and 2ω parts is automatically achieved in the active medium. Our frequency doubling geometric configuration is then completely straightforward and as a result, it can be easily implemented in HH seeding schemes. Moreover, the polarisation of the second harmonic is in this case perpendicular to the polarisation of the fundamental frequency, which corresponds to the situation of most efficient harmonics generation, according to latest results [10]. Finally, due to the group velocity mismatch between ω and 2ω in the non-linear crystal, the IR beam is delayed (18.7 fs for 100 µm thick BBO) compared to the blue beam.

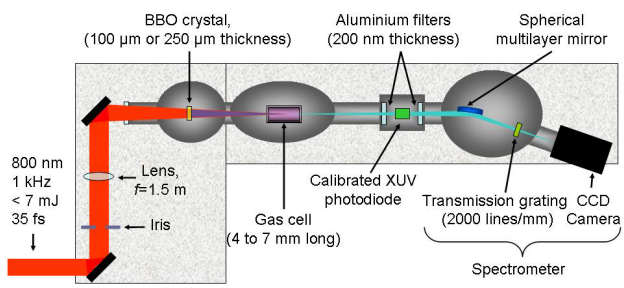

Figure 2: Layout of the kHz two-colour high harmonic generation experiment with a BBO crystal inserted directly in the IR beam path. f is the focal length of the lens used for generating harmonics.

The system for detecting the harmonic intensity content is based on a spectrometer composed of a spherical mirror at grazing incidence (15°) with 800 nm anti-reflection coating (ZrO_2/Si multilayer deposit), a transmission grating (2000 lines/mm with a flat

diffraction efficiency as function of the wavelength in the presented spectral range) and a CCD Camera (Princeton, 1340×400 pixels, 20×20 μm^2). From the recorded pictures (vertical position as a function of the wavelength), spectra are obtained by cuts in the horizontal plan and by the integration of this cut over the vertical dimension. Finally, they can be calibrated in absolute energy per pulse by means of a solarblind XUV photodiode (from NIST laboratory). Two thin aluminium filters (~200 nm thickness each) prevent the IR beam from propagating to the spectrometer, but unfortunately also attenuate the HH photon flux. From 65 nm to 17 nm, the filter transmission assuming pure aluminium is approximately 60 %, but is reduced to a few percents when taking into account oxidation. Below 17 nm, the transmission is close to zero, which gives explanation for the fast decrease of signal observed in the following spectra.

TYPICAL EVOLUTIONS OF SPECTRA

The relatively high energy provided by our kHz IR laser allowed performing a detailed study of a high repetition rate harmonic generation source with two-colour mixing in many gases. The IR peak power densities required for ionisation of xenon, krypton, argon, neon and helium are 0.7, 1, 2, 4 and 7×10^{14} W/cm^2 respectively. For any type of gas, the two-colour effects are visible. To be more precise, Fig. 3, which presents the normalised intensity spectra obtained with Ne and with either ω or ω+2ω technique, perfectly illustrates the three major evolutions: double harmonic content, redshift and increase of efficiency of generation.

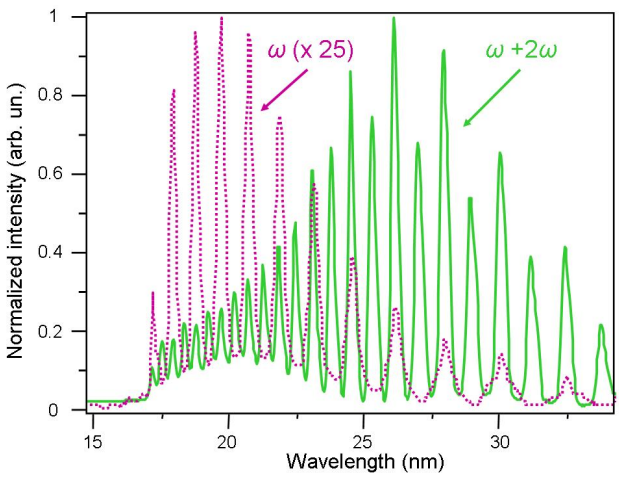

Figure 3: Normalised intensity HH spectra obtained with neon gas with either ω or ω+2ω technique and with same optimisation parameters as for ω: E_ω=6 mJ, Φ=40 mm, L_C=7 mm and P_G=35 mbar. ω (I_ω~7×10^{14} W.cm^{-2}) and ω+2ω (100 μm thick BBO crystal, I_ω~4.5×10^{14} W.cm^{-2} and $I_{2\omega}$~3.4×10^{14} W.cm^{-2}).

High factor of increase is here observed, when keeping the same optimisation parameters as for ω, such as the energy and the IR beam aperture (respectively E_ω and Φ), the gas pressure (P_G) and the cell length (L_C). Spectra present a relatively flat distribution with an intensity ratio from odd to even harmonics close to one, and the intensification arises over the whole spectrum.

Also this enhancement is clearly dependent on the considered gas (Fig. 4), typically He (×~100), Ne (×~25), Ar (×~0.5), Kr (×~0.5) and Xe (×~0.5). As in a two-colour harmonic generation arrangement the efficiency is increased by the higher ionisation rate, it could be also stated that the lower the ionisation rate in the traditional harmonic generation (helium and neon) the stronger the enhancement factor with the mixing. For gases typically rather efficient like argon, krypton and xenon and using the same optimisation parameters as for ω, there could even be actually a decrease of the maximum harmonic signal by a factor of 2. However, as the spectral content is doubled (and as the harmonic spectral width is similar) the whole energy seems to be conserved.

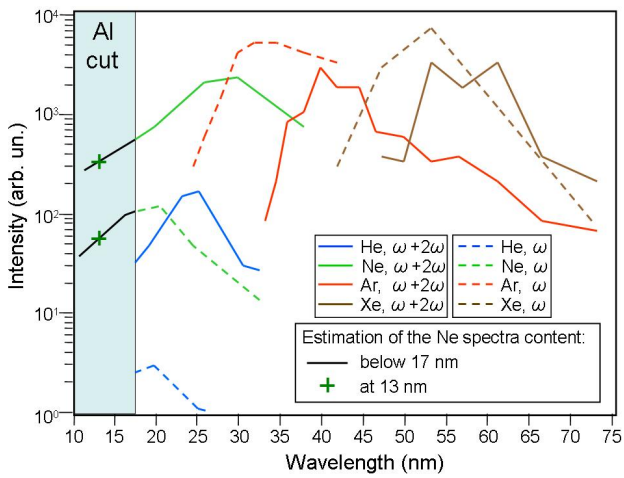

Figure 4: Schematic representation of the achieved experimental intensity for different gases. The curves presented match to the fitted envelope of the recorded spectra. The spectra have been obtained with either ω or ω+2ω technique (100 μm thick BBO crystal) and with same optimisation parameters as for ω: E_ω=6 mJ, L_C=7-9 mm and P_G=30-35 mbar. Xe and Ar cases: Φ=20 mm, I_ω~1.2×10^{14} W.cm^{-2} for ω configuration, and I_ω~0.78×10^{14} W.cm^{-2}, $I_{2\omega}$~0.59×10^{14} W.cm^{-2} for ω+2ω configuration. Ne and He cases: Φ=40 mm, I_ω~7×10^{14} W.cm^{-2} for ω configuration, and I_ω~4.5×10^{14} W.cm^{-2}, $I_{2\omega}$~3.4×10^{14} W.cm^{-2} for ω+2ω configuration. The light-blue window corresponds to the spectral part where the aluminium transmission is close to zero. Then, the black line represents the extrapolation of the extension of the intensity decrease curvature below 17 nm.

Due to this gas type dependence, the effect of flux increase can significantly vary in the observed full spectral range. In other words, the high efficiency of this two-colour process at short wavelengths notably compensates the low efficiency of the classical harmonic generation in this range. Consequently, it extends the spectral region accessible for applications and seeding to shorter wavelengths.

FLUX OPTIMISATION

Since the two-colour harmonic generation process is different from the standard one at the fundamental frequency (short electron path selection and higher ionisation rate), the optimal parameters for both cases have to be notably different. Fig. 5 presents the Ar example. For optimising the flux, both the gas pressure and the cell length are approximately twice smaller. In other words, even with gas like argon relatively efficient in the classical harmonic generation, a reasonable enhancement can be obtained (factor of 2, between the green and red curves), as lower pressure and smaller propagation length in the cell involve smaller absorption in the 40-70 nm range.

Then, in particular conditions, i.e. high intensities for the IR and blue beams, the intensity of this latter being in addition the highest one, spectra can also reveal an additional strong magnification (factor 10, reaching 50 nJ) on some particular orders, i.e. the $2\times(2n+1)$ components of the even orders, while complementary components saturate ($2\times(2n)$). These $2\times(2n+1)$ components are the orders which can be generated by the 2ω component alone, which intensity is stronger in this case.

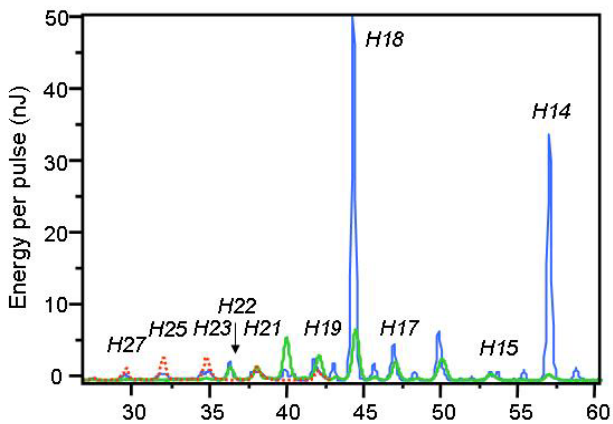

Figure 5: HH spectra calibrated in energy per pulse and obtained in Ar with either ω (dotted line, L_C=8 mm and P_G=30 mbar) or ω+2ω (continuous line, 100 µm thick BBO crystal, L_C=4 mm and P_G=16 mbar) technique, for different laser energies and iris apertures. *H14* to *H27* correspond to the harmonic orders. (a) Full aperture beam (Φ=40 mm). E_ω=6 mJ, I_ω~1.2×10^{14} W.cm^{-2} for ω configuration and E_ω=5.2 mJ, I_ω~3.9×10^{14} W.cm^{-2}, $I_{2\omega}$~2.9×10^{14} W.cm^{-2} for ω+2ω configuration. (b) ω+2ω, E_ω=6.8 mJ and either Φ=40 mm (I_ω~4.4×10^{14} W.cm^{-2}, $I_{2\omega}$~5.4×10^{14} W.cm^{-2}).

Actually, as in a two-colour field harmonic generation configuration, both IR and blue beams have to be intense for reaching the highest HH yield, more driving laser energy is required than in a single-colour configuration for a fixed geometrical configuration of focusing. As a consequence, such an additional effect of yield enhancement on the $2\times(2n+1)$ components of the even orders could not have been observed on gases like He and Ne for which ionisation potentials are higher than for Ar, Kr and Xe., but should be observed in principle in different experimental conditions.

CONCLUSIONS

The two-colour scheme, implemented on our kHz HH line, has allowed us to extensively study both spectral and intensity evolutions of the double content harmonic spectra. Series of spectra have been obtained in the range from 65 nm down to 17 nm using diverse gases for generation. It mainly showed a higher efficiency for He and Ne gases generating at shorter wavelengths and a significant control of the harmonic intensity for specific orders via the gas pressure, the gas cell length, and the intensity of the 2ω component. To be more precise, it has been observed either a limited magnification of the whole spectrum with optimisation parameters close to the ω ones, or a more significant increase of both types of even harmonics with twice smaller cell length and gas pressure, or finally a very strong enhancement mainly located on the $2\times(2n+1)$ components due to a higher 400 nm component. Nevertheless, this final step occurs at higher wavelengths and this is why the adaptation of the ω+2ω technique for seeding experiments requires a strong control of the generation, in order to keep the redshift effect small but still obtain as much as possible photons at the selected wavelength.

To conclude, in order to attain hard X-ray radiations (from 10 nm to 0.01 nm), the use of higher fundamental wavelength lasers, such parametric amplifiers (1.2-1.5 µm [11]), is expected. Indeed, according to equations 1 and 2, it would correspond to a strong blue shift of the cut-off region. Coupled to the two-colour strong increase effect observed in Ne or He gases, intense HH pulses could be generated in such a spectral region of interest for seeding. The so-called water-window (~2-4 nm), vital for the study of biological samples, and perhaps even the Angström region (0.1 nm) could be then reached.

REFERENCES

[1] G. Lambert et al. Nature Physics **889** (4) 296 (2008).
[2] B. W. J McNeil et al. New Journal of Physics **9** 82 (2007).
[3] T. Shintake et al. Nature Photonics **2** 555 (2008).
[4] L. H. Yu et al. Science **289** 932 (2000).
[5] L. Giannessi and P. Musumeci New Journal of Physics **8** 294 (2006).
[6] R. Bonifacio et al. Opt. Commun. **50** (6) 373-378 (1984).
[7] S. Watanabe et al. Phys. Rev. Lett. **73** 2692 (1994).
[8] G. Lambert et al. New Journal of Physics, to be published in 2009.
[9] C. M. Kim et al. Phys. Rev. A **72** 033817 (2005).
[10] I. J. Kim et al. Appl. Phys. Lett. **92** 021125 (2008).
[11] B. Shan et al. Appl. Phys. B **74** 23 (2002).