PRODUCTION OF FEMTOSECOND PULSES IN A FREQUENCY DOUBLER AND PERSPECTIVES OF FLASH USER FACILITY FOR PUMP-PROBE EXPERIMENTS WITH FEMTOSECOND RESOLUTION

E.L. Saldin, E.A. Schneidmiller, and M.V. Yurkov Deutsches Elektronen-Synchrotron (DESY), Hamburg, Germany

Abstract

Free Electron Laser in Hamburg (FLASH) operates successfully since the year of 2000. Permanent upgrades of the facility did allow to reduce operating wavelength from 100 nm in 2000 down to 13 nm in 2006. An upgrade of the year of 2007 is in the progress, and after its completition FLASH will reach design value of the wavelength of 6.4 nm. An attractive feature of FLASH is production of intense, ultra-short radiation pulses of sub-10-fs duration. In this paper we describe perspective upgrades of FLASH aiming at extension of the operating wavelength range while conserving the feature of ultra-short pulse production. We show that an upgrade of FLASH with a frequency doubler will allow to reduce the wavelength down to 3 nm, thus covering the "water window" - the wavelength range that is crucially important for the investigation of biological samples. We show also that recent installations at FLASH of far infrared undulator (FIR) and optical replica synthesizer (ORS) open up the possibility for implementation of schemes allowing to perform pump-probe experiments with 10 femtosecond temporal resolution.

INTRODUCTION

During last years we observe rapid progress of Self-Amplified Spontaneous Emission Free Electron Lasers (SASE FELs) [1–3]. Jump in the wavelength was by about three orders of magnitude, from 12 μ m in 1997 down to 13 nm in 2006 [4–9]. Presently FLASH (Free Electron Laser in Hamburg) has produced unprecedented powers for EUV radiation at a fundamental wavelength of 13.7 nm, and harmonics with wavelengths as low as 2.75 nm [9]. After an energy upgrade to 1 GeV of the FLASH driving linac minimum wavelength in the fundamental harmonic of FLASH will be 6.4 nm.

FLASH demonstrated unique femtosecond mode of operation [6–9] which was not considered at an early design stage of the project [10]. Thorough analysis has shown that due to nonlinear compression and small local energy spread the short high-current leading peak (spike) in the bunch density distribution was produced by beam formation system. Despite strong collective effects (of which the most critical was the longitudinal space charge after compression) this spike was bright enough to drive FEL process up to the saturation for the wavelengths down to 13 nm. Analysis of the latest experimental results [9] indicate

X-ray FELs

that the peak current in the spike is 2 to 2.5 kA, the FWHM length of the high current spike is approximately 30 fs and the normalized emittance is 1 to 1.5 mm-mrad. Note that the latter value is significantly less than the project one of 2 mm-mrad [10], and encouragingly approaches to the values predicted by start-to-end simulations. For FLASH this result is of crucial importance allowing to reach shorter wavelengths with fixed electron energy of 1 GeV. The first scenario for reaching "water window" at FLASH equipped with efficient frequency doubler has been analyzed a few years ago [11, 12]. That analysis has been based purely on refined start-to-end simulations. Now, with an experimentally proven update of the beam parameters we present further development of a concept of frequency doubling for generation of powerful femtosecond pulses. Application of frequency doubler at FLASH will allow to cover the water window (wavelength range between the K-absorption edges of oxygen ($\lambda = 2.34$ nm) and carbon ($\lambda = 4.38$ nm)) that is crucially important for the investigation of biological samples.

Two-color pump-probe experiments are very attractive for time-resolved studies. In this paper we show that present configuration of FLASH holds great potential for pump-probe experiments with 10 femtosecond temporal resolution. This potential stems from recent installation at FLASH of far infrared undulator (FIR) and optical replica synthesizer (ORS) [13–16]. In the case of FIR undulator optical pulses from FIR and XUV pulses from FEL are naturally synchronized since they are produced by the same electron bunch. In the case of pump-probe experiments with an external optical laser an ORS setup is used as a selection trigger for perfectly synchronized pump-probe pulses.

CONCEPT OF FREQUENCY DOUBLER

An idea of using two undulators, with the second undulator resonant to one of the harmonics of the first one, was considered in [17–19] (it is also referred to as the "afterburner" method). The first undulator is long enough to reach saturation and produce strong spatial bunching in harmonics. The bunched beam generates coherent radiation in the second undulator which follows immediately the first one. The main problem with this approach is the large induced energy spread which significantly degrades the performance of the radiator section at the harmonic frequency [11, 12]. Another method to generate higher harmonics is high-gain harmonic generation scheme from the external seed (see [20] and references therein). However, it is technically complicated and is not flexible enough for production of tunable radiation.

The concept of effective frequency doubler scheme for SASE FEL has been proposed in [11, 12]. This scheme essentially exploits the feature of a small local energy spread in the driving electron beam of SASE FEL. It consists of undulator tuned to the first harmonic, dispersion section, and undulator tuned to the second harmonic. The latter one can be also tapered in order to increase output power above saturation level. The first stage is a conventional SASE FEL. The gain of the first stage is controlled in such a way that the maximum energy modulation of the electron beam at the XFEL exit is about equal to the local energy spread, but still far away from saturation. When electron bunch passes through dispersion section this energy modulation leads to effective compression of the particles. Then bunched electron beam enters the 2nd harmonic undulator, and from the very beginning produces strong radiation because of a large value of spatial bunching, low value of the emittance and relatively small induced energy spread. Saturated power has the same level as for conventional SASE FEL, but is reached in significantly shorter undulator.

PRODUCTION OF ELECTRON BUNCHES

We illustrate operation of the frequency doubler for the energy of the driving beam of 1 GeV (see Fig. 1). The electron beam is produced in a radio frequency gun and brought up to an energy of 1000 MeV by six accelerating modules ACC1 to ACC6. At energies of 130 and 380 MeV the electron bunches are compressed in the bunch compressors BC1 and BC2. The electron beam formation system is based on the use of nonlinear longitudinal compression. When the bunch is accelerated off-crest in the accelerating module, the longitudinal phase space acquires a radio frequency induced curvature. Downstream of each bunch compressor, this distortion results in a non-Gaussian distribution within the bunch and in a local charge concentration. It is the leading edge of the bunch, with its high peak cur-



Figure 1: Schematic layout of the FLASH facility equipped with frequency doubler. Undulator of frequency doubler is placed just after the main undulator. Abbreviations ACC, BC, and DS stand for accelerating module, bunch compressor, and dispersion section, respectively. The scheme also shows far infrared undulator (FIR) and optical replica synthesizer (ORS) setups installed at FLASH in 2007.



Figure 2: Structure of the electron bunches at FLASH: current I (solid line) and mean energy E (dashed line) along the electron bunch at the undulator entrance (simulations of [9] scaled to 1 GeV). The bunch head is located at the right hand side of the figure.

rent, which is capable of driving the high intensity lasing process (see Fig. 2). Collective effects play a significant role in the bunch compression process for short pulses. One can see on Fig. 2 that an increase in the peak current and a narrowing of the width of the spike leads to an increase in the induced energy chirp due to space charge effects.

PROPERTIES OF THE RADIATION

We illustrate operation of the frequency doubler at FLASH for the case of FEL process driven by short-spike electron bunch with energy of 1 GeV (see Fig. 2). The main undulator of FLASH is a fixed 12 mm gap permanent magnet device with a period length of 2.73 cm and a peak magnetic field of 0.48 T. The frequency doubler is 9 m long undulator (1.95 cm period and 0.39 T peak magnetic field). Simulations have been performed with time-dependent code FAST [21] upgraded for simulation of higher harmonics. Prior presenting results for the frequency doubler it is worthwhile to overview operation of SASE FEL with a uniform undulator. Simulations of the femtosecond mode of operation of FLASH at 6 nm show that present undulator length of 27 meters is sufficient to reach deep saturation in the main undulator. Expected level of the average energy in the radiation pulse is up to 70 μ J for an ideal tuning of the machine and emittance in the bunch head of 1.4 mm-mrad. Expected radiation pulse duration is in sub-10 fs range.

Electron bunch quality is high enough to drive SASE FEL optimized for production of 3 nm radiation (see Fig. 3). Saturation is achieved at the undulator length of about 30 meters with the level of the energy in the radiation pulse about 15 μ J. We present this number for the further comparison with the frequency doubler.

When operating as a frequency doubler, the level of output radiation energy in the main undulator should be tuned



Figure 3: Energy in the radiation pulse in the frequency doubler (solid line). Electron energy is 1 GeV, undulator period is 1.95 cm, and peak magnetic field is 0.39 T. Radiation wavelength is 3.2 nm. Dashed line shows operation of SASE FEL with the same undulator.

to the value about one microjoule. At this stage of amplification process induced energy modulations become to be about local energy spread in the lasing spike. Then electron bunch passes dispersion section with the net compaction factor about one micrometer, and energy modulations transforms to the density modulation with high content of the second harmonic. Finally, electron bunch is directed to the undulator resonant at the second harmonic (frequency doubler undulator) and readily starts to produce powerful radiation from the very beginning. Since the electron beam still has high quality, a self-consistent process of radiation amplification takes place. Peak radiation intensity is nearly the same as in conventional SASE FEL, but saturation occurs at a shorter undulator length. We see from Fig. 3 that the level of the average radiation energy is 5 μ J at the undulator length of 9 meters. Temporal and spectral properties of the radiation pulse at this point are illustrated with Fig. 4. FWHM pulse length is well below 10 fs, and average power exceeds gigawatt level. Contribution of the 3rd harmonic into the full radiation power is about 0.08%, and peak power is in a megawatt range.

PERSPECTIVES FOR PUMP-PROBE EXPERIMENTS ON A 10-FS TIME SCALE

Recently FLASH facility has been extended with the far infrared undulator (FIR) and optical replica synthesizer (ORS) [13–16]. These devices hold great potential for organization of pump-probe experiments on a 10-fs pulse scale at FLASH operating in a femtosecond mode.

Current plans for FIR undulator foreseen pump-probe experiments with VUV and FIR pulses (wavelength range from a few to 200 μ m) [14]. While initially FIR and VUV pulses are synchronized on a 10 fs timescale (they both are produced by the same electron bunch), additional jit-



Figure 4: Temporal (top) and spectral (bottom) properties of the radiation pulse at the length of the frequency doubler of 9 meters. Bold lines show averaged values, and thin lines are single shots.

ter may be introduced by mechanical vibrations of optical components because different beamlines are used for transport of VUV and FIR pulses. Here we propose jitter-free scheme (see Fig. 5). Combination of modulator of ORS and FIR undulator as a radiator will allow to produce powerful pulses of coherent radiation with the wavelength of driving laser around one micron. Then both pulses are transported to the experimental area using the VUV opti-



Figure 5: Scheme for pump-probe experiments with VUV FEL pulses and pulses produced by infrared undulator. Operation of the scheme is based on production by the same electron bunch of naturally synchronized VUV FEL pulses and pulses produced by the infrared undulator. Relative synchronization of the pulses is preserved by means of using the same optical system for transportation of the pulses to the experiment.



Figure 6: Scheme for pump-probe experiments with VUV FEL pulses and optical pulses from external femtosecond laser (top). ORS setup driven by the same external laser produces selection trigger for well synchronized pulses. Bottom plot illustrates overlapping of electron pulse (solid line) and optical pulse (dashed line) in the modulator of ORS setup.

cal system which conserves mutual synchronization. Calculations of the photon beam transport show that both radiation pulses, VUV and infrared one, can be effectively transported through existing optical system. Separation of infrared and VUV pulses and organization of required time delay is performed just near the experimental sample.

ORS setup can be also used for organization of pumpprobe experiments involving femtosecond pulses from VUV FEL and powerful optical pulses from femtosecond optical laser. Key problem of such experiments is precise synchronization of laser and electron pulses having timing jitters of different physical nature. Instead of solving synchronization problem we propose to use pulse selection scheme based on ORS setup. Operation of this scheme is illustrated in Fig. 6. An optical beam from fs-laser is split into two beams, one is directed to the sample, and another one is used as a seed for ORS. Output signal from the ORS exhibits strong dependence on the time jitter between the lasing spike and short laser pulse, and achieves maximum value at perfect overlapping. Selection signal is generated in this case which marks events with perfectly synchronized pump-probe pulses.

Installation of frequency doubler at FLASH is natural, cost-effective way for complete covering of the "water window", the wavelength range which is of crucial importance for studying biological samples. But it is not just simple extension of the operating wavelength range of the facility: production of ultra-short pulses with gigawatt level of output power extends an opportunity for studies of timedependent processes on a sub-10-fs time scale described above. In addition, the frequency doubler setup produces two radiation pulses of different frequencies which are naturally synchronized on a few femtosecond scale. The frequency doubler scheme is rather flexible allowing to negotiate with the intensity of the radiation pulses. In particular, it is rather simple to produce VUV pulses (with fundamental and doubled frequencies) with similar intensity in the range of a few microjoules which reveals an opportunity for performing pump-probe experiments with powerful VUV pulses of different colors.

ACKNOWLEDGEMENT

We thank R. Brinkmann, J. Rossbach, and J.R. Schneider for interest in this work.

REFERENCES

- A.M. Kondratenko and E.L. Saldin, Part. Accelerators 10 (1980)207.
- [2] Ya.S. Derbenev, A.M. Kondratenko, and E.L. Saldin, Nucl. Instrum. and Methods 193(1982)415.
- [3] J.B. Murphy and C. Pellegrini, Nucl. Instrum. and Methods A 237(1985)159.
- [4] M. Hogan et al., Phys. Rev. Lett. 81(1998)4867.
- [5] S.V. Milton et al., Science **292**(2001)2037.
- [6] V. Ayvazyan et al., Phys. Rev. Lett. 88(2002)104802.
- [7] V. Ayvazyan et al., Eur. Phys. J. D 20(2002)149.
- [8] V. Ayvazyan et al., Eur. Phys. J. D 37(2006)297.
- [9] W. Ackermann et al., Nature Photonics, 1 (2007)336.
- [10] T. Åberg, et al., A VUV FEL at the TESLA Test Facility at DESY, Conceptual Design Report, DESY Print TESLA-FEL 95-03, May 1995.
- [11] J. Feldhaus et al., DESY Print DESY 03-092, July 2003.
- [12] J. Feldhaus et al., NIM A528(2004)471.
- [13] B. Faatz et al., Nucl. Instrum. and Methods A475(2001)363.
- [14] M. Gensch et al., "New THz undulator beamline at the VUV FEL FLASH", submitted to Infrared Physics & Technology.
- [15] E.L. Saldin, E.A. Schneidmiller and M.V. Yurkov, Nucl. Instrum. and Methods A539(2005)499.
- [16] P. van der Meulen et al., Proc. of FEL 2006 Conference, Berlin, 2006, p. 296.
- [17] R. Bonifacio, L. De Salvo, and P. Pierini, Nucl. Instrum. Methods Phys. Res. A 293, 627(1990)
- [18] Ciocci, et al., IEEE J. Quantum Electron. 31 (1995)1242
- [19] W. M. Fawley et al., Proceedings of the IEEE 1995 Partice Accelerator Conference, 1996, p219
- [20] I. Ben-Zvi et al., Nucl. Instrum. Meth. A304(1991)151.
- [21] E.L. Saldin, E.A. Schneidmiller, and M.V. Yurkov, Nucl. Instrum. and Methods A 429(1999)233.