OPERATION OF THE FIRST UNDULATOR-BASED FEMTOSLICING SOURCE*

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Abstract

At the BESSY II storage ring, a source of sub-100 fs x-ray pulses with tunable polarization and excellent signalto-background ratio has been constructed in 2004, based on laser-induced energy modulation ("femtoslicing") and subsequent angular separation of the short-pulse x-rays from an elliptical undulator. Ultrashort x-ray pulses are now routinely delivered for pump-probe applications.

INTRODUCTION

The study of the structure and function of matter with synchrotron radiation at short wavelengths is limited in time resolution by the electron bunch length in synchrotron light sources to 30-100ps (fwhm). Mode-locked lasers with chirped-pulse amplification, on the other hand, have reached the 100-fs regime at visible wavelengths in the mid-1980s [1]. "Femtoslicing" is a method to obtain short pulses and short wavelength. Even though the photon flux is low $(10^6-10^7 \text{ photons per s and } 0.1\% \text{ bandwidth})$, this short-pulse technique is currently without competition at photon energies above a few 100 eV, a regime not yet reached by free-electron lasers and in which the flux from laser harmonics or laser-based plasma sources is even lower. The only source with comparable properties was the SPPS [2] at SLAC, which has by now discontinued its operation.

The femtoslicing technique was proposed [3] and experimentally demonstrated [4] at the Advanced Light Source (ALS) in Berkeley producing 150-fs x-ray pulses from a bend magnet. A first undulator-based femtoslicing source was constructed in 2004 at BESSY in Berlin [5] and is now in routine operation for pump-probe applications. In 2006, another undulator-based source is commissioned at the ALS [6] and a third one has been completed at the Swiss Light Source (SLS) in Villigen [7].

PRINCIPLE OF OPERATION

The operation principle is sketched together with the BESSY installation in Fig. 1. A femtosecond laser pulse copropagates with a bunch of 1.7-GeV electrons (Lorentz factor $\gamma \sim 3300$) in an undulator (the "modulator" U139 with 10 periods of length $\lambda_{\rm U} = 139$ mm), where the energy of electrons in the overlap region is modulated with



Figure 1: BESSY II floorplan (top) and schematic view (bottom) of modulator (U139), radiator (UE56) and a bend magnet (B2) for angular short-pulse separation. The main pulse (red solid line) from a Ti:sapphire amplifier (amp) is focussed by a telescope (T) into the modulator and is monitored at a diagnostics station (D), while a 10%-fraction is sent as pump pulse (dashed red line) to the experiment at the x-ray beamline (blue). The interaction efficiency is monitored by coherent THz radiation (magenta).

a periodicity equal to the laser wavelength $\lambda_{\rm L} = 780$ nm. The undulator parameter K is tuned to match the resonance condition $\lambda_{\rm L} = \lambda_{\rm U} (1 + K^2/2)/2\gamma^2$. Depending on the pulse energy (typically 2 mJ) and pulse duration (40-50 fs), the energy modulation can reach 1% of the beam energy. The laser oscillator operates at 83.3 MHz (1/6 of the storage ring rf), the amplifier at 1 kHz [8]. Off-energy electrons are displaced in horizontal angle by a dipole magnet (bend angle 112 mrad) and their radiation in a subsequent elliptical undulator (the "radiatior" UE56 with 30 periods of length 56 mm and tunable elliptical polarisation) can be extracted using an aperture at a certain angle with respect to the electron beam axis. Even though this short-pulse radiation is only 10^{-4} of the radiation from the whole bunch, the angular separation scheme [4] allows to obtain a signal-tobackground ratio of 10-100. In contrast to that, separation by parallel electron displacement with subsequent imaging of their radiation on a slit suffers from severe background due to non-specular scattering from the focusing elements

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Figure 2: Electron loss rate versus scraper position with (dots) and without (open symbols) energy modulation under variation of the laser pulse energy. The solid lines are simulation results. Inset: modulation amplitude $\Delta E/E$ versus applied (dots) and simulated (squares) pulse energy. Error bars reflect the uncertainty in the optical functions.

[4]. Note that the use of an undulator as radiator not only leads to orders of magnitude more photon flux per bandwidth, compared to a bend magnet, but is a prerequisite to angular separation and to optimum background rejection. The x-ray pulse duration can be optimized by minimizing the non-isochronicity of electrons between modulator and radiatior. In the BESSY case, modulator and radiator are placed in the same straight section with only one dipole magnet between them. The pulse length is mainly determined by the laser pulse duration and the R_{51} matrix element (path length per horizontal offset) of that magnet, and amounts to 100 fs (fwhm). The radiator is optimized for photon energies up to 1400 eV, covering e.g. the K-edges of N and O, the L-edges of Fe, Co and Ni, and the M-edges of rare-earth elements. It can be tuned from horizontally linear to circular and vertically linear polarization, particularly in view of magnetic studies. For pump-probe applications, a 10%-fraction of the laser pulses is sent through an evacuated pipe to the x-ray beamline, which comprises a plane-grating monochromator [9]. These pump pulses are intrinsically synchronized with the short x-ray pulses and their timing is controlled on the fs level by a motorized delay stage. X-ray signals are detected by an avalanche photodiode (APD), either by recording them with a digital oscilloscope or counting pulses that exceed a disciminator threshold, while the pump pulses are sufficiently attenuated by the sample under study. A chopper wheel blocks every other pump pulse in order to suppress systematic errors from drifts of the short-pulse intensity.

ENERGY MODULATION

Laser and U139 radiation are continuously monitored via water-cooled Cu mirrors at a diagnostics station (labelled D in Fig. 1), comprising an APD to verify correct laser-electron timing and CCD cameras to observe the transverse overlap with two different foci. A few meters



Figure 3: Top – angular distributions of photon signals at 1200 eV, 850 eV and 400 eV with (solid dots) and without laser interaction (squares) and 1 ms after the interaction (open dots). Bottom – ratio of signal and laser-off background (squares) or background after 1 ms (open dots).

downstream of the modulator, the non-isochronicity of offenergy electrons causes a dip and two side lobes in the longitudinal electron distribution, which persist over several turns and give rise to coherent radiation the THz regime [10]. Once the laser-electron overlap has been established, which is usually accomplished within minutes, THz radiation is used to optimize and maintain the laser-electron interaction by feedback loops.

In dedicated shifts, the energy modulation has been quantified by experiments, in which a horizontal scraper was moved towards the electron beam while recording the electron loss rate with and without laser interaction [11]. While the transverse position is proportional to the amplitude of energy modulation $\Delta E/E$, the loss rate measures the number of electrons exceeding an amplitude corresponding to the scraper position. Results under variation of the laser pulse energy are shown in Fig. 2, following the theoretical expectation of $\Delta E/E \sim \sqrt{E_{\rm L}}$ [3]. Simulations, however, obtain the same energy modulation with about half the applied pulse energy. This discrepancy (also observed in [4]) is attributed to optical aberrations and requires futher investigation.

ULTRASHORT X-RAY PULSES

Based on earlier measurements of the UE56 angular distribution over seven orders of magnitude, a signal-tobackground ratio of 10 was predicted [12]. Fig. 3 shows recently measured angular distributions with and without energy modulation for different photon energies. Here, an APD signal of 1 V corresponds to $\sim 10^3$ photons/(s 0.1% bandwidth). Also shown is the background from electrons excited by the previous laser interaction. Their signal, normalized to the short-pulse signal at time zero, is plotted in Fig. 4 on a logarithmic time scale. A 200-kHz modulation corresponds to the aliased betatron frequency $f_{\circ}(1-q)$ with revolution frequency $f_{\circ} = 1.25$ MHz and fractional



Figure 4: APD signal of 850-eV photons normalized to the short-pulse signal versus time after the laser interaction. Up to 1 ms, a data point is shown for every revolution $(0.8 \,\mu s)$.

tune q = 0.84, and a 14-kHz modulation to twice the synchrotron frequency. After 1 ms, the betatron oscillation has randomized, leaving a structureless background with an exponential decay time of 5 ms. At the present interaction rate of 1 kHz, it is advantageous to accept this background and use a single bunch of 5-10 mA, placed in the ion-clearing gap of the 250-mA multi-bunch fill (0.7 mA per bunch). Upgrading the laser system to higher repetition rate will require to interact with several bunches in turn.

For circular polarization, additional background from radiation with the opposite helicity must be considered. This is due to the fact that the circular polarization component of an elliptical undulator changes sign for a sufficiently large angle (here 0.2 mrad) between electron and photon.

The spectral distribution of short-pulse photons and their background is shown in Fig. 5. Here, the monochromator was set to 1200 eV and the magnetic UE56 gap was varied, exhibiting the undulator spectrum from the 3rd to the 9th harmonic. For a fixed separation angle, the angular distribution becomes wider with decreasing gap (increasing *K*) and the background rises, which can be counteracted by increasing the angle.

Indications of the expected pulse duration of 100 fs are (i) the THz spectrum, which is compatible with the expected longitudinal electron distribution at the THz beamline [10], and (ii) recent pump-probe experiments (to be published) suggesting a time resolution of 150 fs including possible path length drifts over 8 hours. It is noteworthy that a 10^{-6} change between the respective paths of the pump and the probe pulses, e.g. through thermal expansion, corresponds to more than 100 fs delay.

OUTLOOK

Presently, short x-ray pulses are generated at a rate of 1 kHz and with $\sim 10^3$ photons per pulse and 0.1% bandwidth. The beamline reduces this flux by two orders of magnitude, the detector efficiency accounts for another factor of 5-10. The rate of detected photons can be increased by raising the laser repetition rate, by improving the beam-



Figure 5: APD signal of 1200-eV photons as a function of the UE56 magnetic gap with and without laser interaction, as well as 1 ms after the interaction.

line transmission, and by using a detector array to detect photons of different energy simultaneously and with better efficiency. Each of these measures can reduce the measurement time by one order of magnitude, thus increasing the range of possible experiments with this novel source.

It has been pointed out that the temporal profile of the laser pulses can be controlled [13] in order to influence the shape of x-ray pulses and of THz pulses [14], e.g. creating double pulses, pulse trains or arbitrary shapes within the resolution limited by laser-electron slippage and other lengthening effects. Double pulses may be useful for THz-THz pump-probe experiments, where the laser pulse energy determines the spectral characteristics of these pulses. A weak laser pulse followed by a stronger one may be used to generate a THz pump and x-ray probe pulse. Coherently radiating pulse trains would enhance a particular wavenumber [14].

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