COHERENT HARMONIC GENERATION AT THE DELTA STORAGE RING: TOWARDS USER OPERATION*

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Abstract

At DELTA, a 1.5-GeV synchrotron light source at the TU Dortmund University, a short-pulse facility based on Coherent Harmonic Generation (CHG) is in operation and shall soon be used for pump-probe experiments. Due to the interaction of ultrashort laser pulses with electron bunches in an undulator, CHG provides short and coherent pulses at harmonics of the laser wavelength. In this paper, recent progress towards user operation, pulse characterization studies such as transverse and longitudinal coherence measurements as well as CHG in the presence of an RF phase modulation are presented.

INTRODUCTION

High-gain free-electron lasers (FELs) are almost ideal radiation sources to study the structure and function of matter, combining short wavelength with femtosecond pulse duration and extremely high peak brilliance. However, to date only four of these machines are in user operation (FLASH, LCLS, SACLA, and FERMI), and being based on linear accelerators, their pulse repetition rate is low and they serve only one experiment at a time. In contrast to that, the pulse duration at synchrotron light sources based on storage rings is 30 to 100 ps, given by the bunch length, but there are about 50 of these facilities in operation [1], each providing up to 40 beamlines simultaneously with soft and hard x-rays at a rate of up to 500 MHz. For experiments that do not require or cannot even tolerate the high peak intensity delivered by high-gain FELs, methods to reduce the pulse length can significantly extend the scientific opportunities of conventional synchrotron light sources.

At synchrotron light sources, pulses in the femtosecond regime can be obtained by separating radiation from a small longitudinal part of the electrons from the rest. Such a 'slice' is defined by the interaction of electrons with a co-propagating ultrashort laser pulse in an undulator (the 'modulator') leading to a periodic modulation of the electron energy within the slice. In the case of incoherent radiation from a subsequent undulator (the 'radiator'), a spatial separation of the short radiation component is required, a method known as femtoslicing [2–5].

Coherent Harmonic Generation

Another method based on the interaction of an ultrashort laser pulse with an electron bunch is called coherent harmonic generation (CHG) [6–9]. Here, the energy modulation

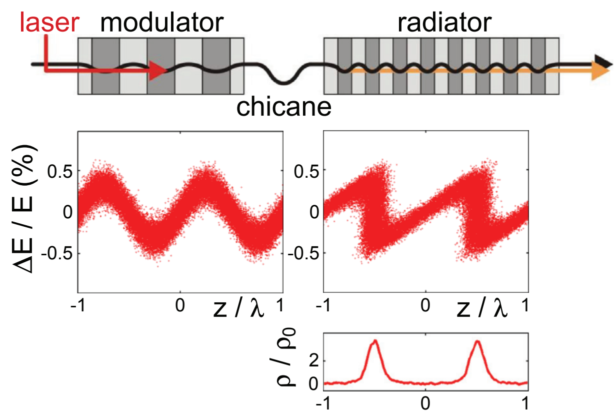

Figure 1: Sketch of the magnetic setup for CHG (top), the longitudinal phase space before and after the magnetic chicane (center), and the longitudinal electron density after the chicane (bottom).

is converted into a density modulation ('micro-bunching') using a magnetic chicane. This leads to the emission of a short coherent pulse in the radiator at harmonics of the laser wavelength. A sketch of this technique is depicted in Fig. 1.

Due to the coherent nature of this radiation mechanism, the CHG pulse can be more intense than the incoherent radiation from the rest of the bunch, such that no spatial separation is required. The radiated power is given by

$$P_{\text{inc}} = N_e \cdot P_e, \tag{1}$$

$$P_{\text{coh}} = \left(\frac{\tau_L}{\tau_b} \cdot N_e\right)^2 \cdot b_n^2 \cdot P_e, \tag{2}$$

where P_{inc} is the incoherent power emitted by an electron bunch with N_e electrons, P_e is the power emitted by a single electron, and P_{coh} is the CHG power emitted by the laser-modulated slice. The number of electrons contributing to the coherent emission is given by N_e times the ratio of laser pulse length τ_L and electron bunch length τ_b. The so-called bunching factor b_n is a measure for the degree of micro-bunching with a value between 0 and 1. It is given by [10]

$$b_n = e^{-\frac{1}{2}n^2 \cdot B^2} \cdot J_n(n \cdot A \cdot B), \tag{3}$$

with

$$A = \frac{\Delta E}{\sigma_E} \quad \text{and} \quad B = r_{56} \cdot \frac{2\pi}{\lambda_L} \cdot \frac{\sigma_E}{E},$$

and the harmonic number n of the laser wavelength λ_L to which the radiator is tuned. Here, J_n is the Bessel function

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of the order n, ΔE is the amplitude of the energy modulation, E is the beam energy, σ_E is the natural energy spread of the electron bunch, and the transfer matrix element r_{56} is the strength parameter of the magnetic chicane.

With a typical bunching factor of the order of $b_n \approx 0.1$, a fraction of contributing electrons of $\tau_L/\tau_b \approx 10^{-3}$, and $N_e \approx 10^{10}$, this leads to a power ratio of $P_{\mathrm{coh}}/P_{\mathrm{inc}} \approx 100$.

In contrast to femtoslicing, which suffers from an extremely low photon rate due to incoherent radiation from a small fraction of the bunch, the intensity of the CHG pulses is large. However, the wavelength is restricted to harmonics of the incident seed wavelength, and due to the dependence $\sim e^{-n^2}$ of the bunching factor, this method is only effective up to about $n = 5$. Instead of using the fundamental wavelength of a femtosecond laser (usually 800 nm from a titanium:sapphire laser system), frequency-doubled or -tripled laser pulses may be used for seeding in order to reach shorter CHG wavelengths.

In 2009, a new FEL seeding scheme was proposed, known as echo-enabled harmonic generation (EEHG) [11], in which a twofold laser-electron interaction leads to significant bunching at much higher harmonics. An implementation of the EEHG scheme to generate ultrashort coherent pulses is planned at DELTA [12].

SETUP AT DELTA

Following proof-of-principle experiments in the 1980s, e.g. [13], recent implementations of the CHG principle were undertaken at ELETTRA [8] in Trieste, Italy, at UVSOR [9] in Okazaki, Japan, and at DELTA [14, 15] in Dortmund, Germany.

DELTA is a 1.5-GeV electron storage ring operated as a light source by the Center for Synchrotron Radiation at the TU Dortmund University. In 2011, a short-pulse facility for coherent VUV and THz pulses based on the CHG principle was installed in the northern section of the storage ring (Fig. 2) [16, 17]. In contrast to other storage rings, the CHG facility at DELTA is fully compatibility with standard user operation due to seeding at the nominal beam energy, and up to 50 days of dedicated machine time are available per year for characterization and optimization of the CHG source.

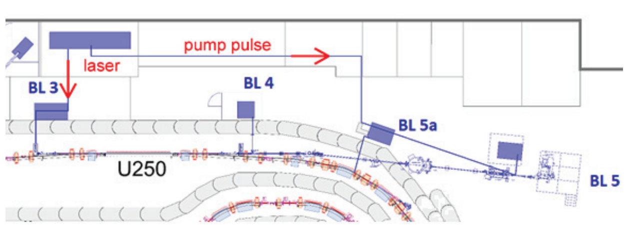

Figure 2: Sketch of the short-pulse facility in the northern section of the DELTA storage ring, including the seeding beamline BL 3, the electromagnetic undulator U250, the diagnostics beamline BL 4, the VUV beamline BL 5, and the THz beamline BL 5a (see text for details).

Laser System and Seeding Beamline

The seeding beamline BL3 features a titanium:sapphire laser system, which generates pulses with a duration of 40 fs (FWHM), a wavelength of 800 nm, and a pulse energy of 8 mJ at a repetition rate of 1 kHz. Using a second- and third-harmonic-generation unit (SHG, THG), the seeding wavelength can be converted to 400 nm or 267 nm, respectively. An optical parametric amplifier can be employed to continuously adjust the seeding wavelength between 470 nm and 1150 nm.

Thus far, seeding is performed with the fundamental wavelength of 800 nm and with 400 nm from the SHG unit. The 800-nm pulses are focused with a set of three lenses, two of which are motorized to adjust position and size of the laser waist in the modulator independently. In the case of 400 nm, the focusing is performed using one movable and one fixed curved mirror. Here, position and size of the waist are correlated.

A fraction of the focused beam is reflected back into the laser laboratory, where a 'virtual beam waist' is observed and optimized at the same distance from the telescopes as the actual modulator.

In future, also the third harmonic of the laser wavelength will be employed for seeding, and a common beam path will be used for all wavelengths (Fig. 3). For focusing, two consecutive telescopes with two curved mirrors each will be installed. Replacing lenses by curved mirrors will reduce pulse lengthening and self-phase modulation. Furthermore, a larger section of the beam path will be in vacuum to reduce pulse distortion in air, and therefore stabilize the CHG intensity. The first telescope (in air) increases the spot size on the vacuum window, while the second telescope (in vacuum) focuses the beam and both together allow to adjust waist size and position independently. Due to the wavelength-dependence of the Rayleigh length, the waist radius and the beam divergence cannot be both the same for each seeding wavelength. A solution has been found to achieve satisfactory waist parameters for all wavelengths using the same dielectric mirrors with a multi-wavelength coating.

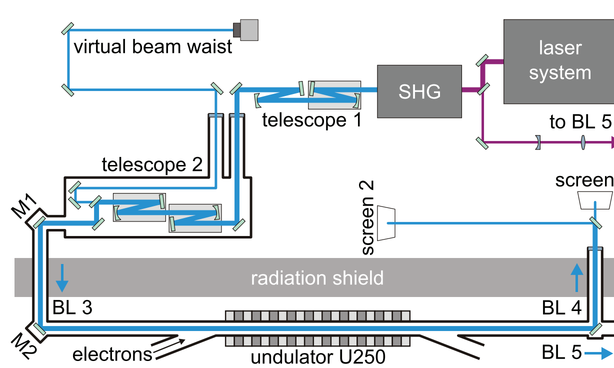

Figure 3: Sketch of the laser beam path, including the planned multi-wavelength reflective telescopes (see text for details).

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The remotely controlled mirrors M1 and M2 (Fig. 3) and a digital RF phase shifter (vector modulator) allow to adjust the transverse and longitudinal overlap of the laser pulses and electron bunches.

Optical Klystron and Diagnostics Beamlines

The electromagnetic undulator U250 (Fig. 2) serves as modulator, dispersive chicane, and radiator by using three independent power supplies.

In-air diagnostics and characterization experiments are performed at the diagnostics beamline BL 4, while radiation with a wavelength below 200 nm is studied at the VUV beamline BL 5 operated by Forschungszentrum Jülich.

The diagnostics beamline BL 4 features fast photodiodes and a streak camera to optimize the temporal laser-electron overlap, as well as CCD cameras observing undulator and laser light at different distances from the modulator to optimize the transverse overlap. Characterization studies of the CHG radiation are performed using power meters, photodiodes, CCD spectrometers, a Czerny-Turner-type monochromator equipped with an avalanche photodiode (APD), and a fast-gated iCCD camera[1].

Following the laser-electron interaction in the undulator U250, the off-energy electrons travel on dispersive orbits through the subsequent magnets. Thus, path length differences lead to the formation of a sub-ps dip in the longitudinal electron density giving rise to coherent THz radiation pulses, which are extracted by the dedicated THz beamline BL 5a [17]. Besides serving as diagnostics for the laser-electron overlap, the THz beamline is used for machine studies [18] and offers intense and short pulses for time-resolved THz spectroscopy.

Pump-Pulse Beamline and User Experiments

For future pump-probe experiments, a fraction of each laser pulse is guided directly to the VUV beamline BL5. Here, the pulses are focused on the sample and transversely stabilized using a feedback system based on position-sensitive diodes and piezo-driven mirrors. The delay between pump and probe pulse is controlled with fs precision by mirrors on a motorized linear stage.

The transverse and temporal overlap between pump and probe pulses on the sample has been established. A first photoemission pump-probe experiment studying the phenomena in ferromagnetic systems using the linear magnetic dichroism [19] is under preparation.

EXPERIMENTAL RESULTS

Characterization of the CHG Pulses

In good agreement with Eqs. 1 and 2, the measured CHG radiation at the second harmonic of the seeding wavelength is up to 600 times brighter than the incoherent radiation, while the third harmonic is still 150 times brighter [14]. With a seeding wavelength of 400 nm, the fifth harmonic

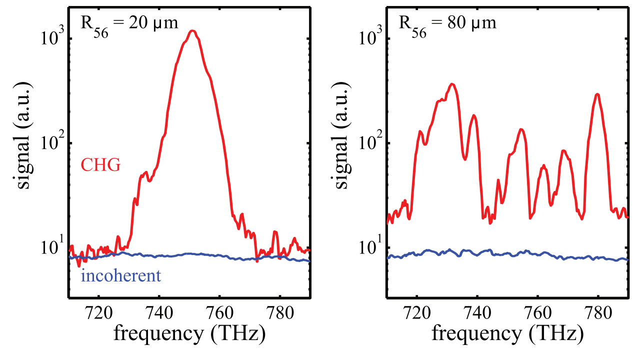

Figure 4: Spectra of CHG and spontaneous undulator radiation around 750 THz (wavelength 400 nm, second harmonic of 800 nm) measured using a Czerny-Turner monochromator with an APD as detector. With a chicane strength far above the optimum value (right), interference fringes in the CHG spectrum emerge [15].

(80 nm) could be observed, and during 800 nm seeding, the seventh harmonic (114 nm) was detected so far.

While seeding with 800 nm and 400 nm, the CHG spectra have been studied extensively under variation of several parameters such as the chicane strength (Fig. 4) or the chirp of the seed laser pulse. CHG spectra are measured with a linear CCD spectrometer, a Czerny-Turner monochromator with an APD, as well as by scanning the monochromator at the VUV beamline BL5. All measurements with optimized chicane strength (e.g. left part of Fig. 4) show a similar spectral width close to the Fourier limit [14].

The coherence of the laser-induced radiation has been studied both transversely and longitudinally [15]. The transverse coherence is measured using a classical double-slit experiment with variable slit separation and a fast-gated iCCD camera as detector. Given by the decreasing visibility of the interference fringes with increasing slit separation, coherence lengths of 1.5 mm and 0.8 mm for 400- and 200-nm

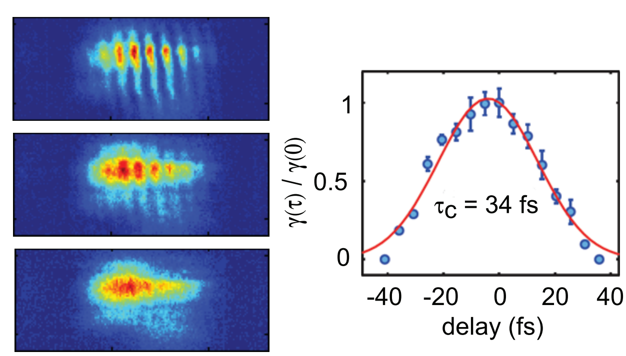

Figure 5: Interference patterns (left) of CHG pulses recorded with a fast-gated iCCD camera behind a double-slit. Using a combination of fused-silica wedges, a relative delay (increasing from top to bottom) between the light from both slits was introduced, leading to a decrease of the fringe visibility γ (right) [15], which allows to calculate of the longitudinal coherence time τ_c.

[1] The camera was generously provided by B. Schmidt and S. Wunderlich, DESY, Hamburg.

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CHG radiation (seeding wavelength of 800 nm) have been measured. As part of an ongoing collaboration with the University of Siegen, the transverse coherence is also studied by analyzing speckle patterns generated by single CHG pulses scattered from a thin organic film [20].

The coherence time, or temporal coherence length, has been studied using both a Michelson interferometer and a double slit experiment, in which the light from one of the slits was delayed using a combination of fused-silica wedges [15]. First consistent results obtained with both setups yield a coherence time of 34 fs (see Fig 5), while the coherence time of spontaneous undulator radiation is only 10 fs partly given by a bandpass filter.

CHG in the Presence of an RF Phase Modulation

During standard user operation at DELTA, a phase modulation of the accelerating RF with twice the synchrotron frequency is routinely applied. The theory of RF phase modulation is described e.g. in [21]. Depending on the amplitude of the modulation, this leads to a longitudinal ‘breathing’ of the electron bunches (Fig. 6, center) or to the formation of two separate stable islands, which rotate in longitudinal phase (Fig. 6, right). As a consequence, the average electron density is reduced and the beam lifetime increases.

In general, the varying electron density would lead to a decreased CHG intensity with large fluctuations. However, adjusting the modulation frequency to a value close to a multiple of the laser repetition rate, the laser-electron interaction samples different phases of the bunch length oscillation at a slow rate of typically 2π per minute, and therefore the laser-induced CHG and THz signals exhibit a beating behaviour as shown in Fig. 7.

Surprisingly, the THz and CHG signals oscillate with 180 degree phase difference. This can be explained by assuming that the CHG signal primarily depends on the number of participating electrons according to Eq. 2 with the exponential factor in Eq. 3 being close to unity, while the THz signal

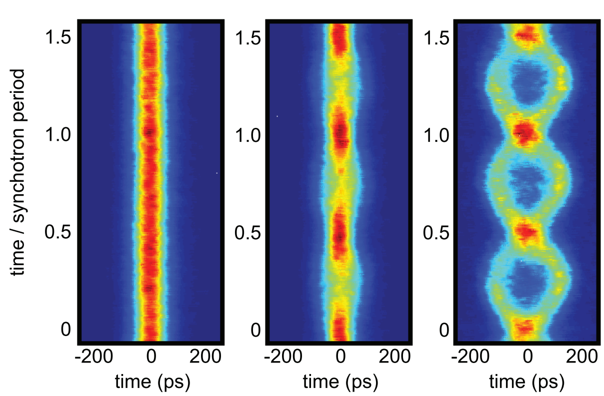

Figure 6: Streak camera images showing the evolution of the longitudinal bunch profile (horizontal axis) over 1.5 synchrotron periods (vertical axis) for an undisturbed beam (left) and in the presence of an RF phase modulation with increasing amplitude (center and right).

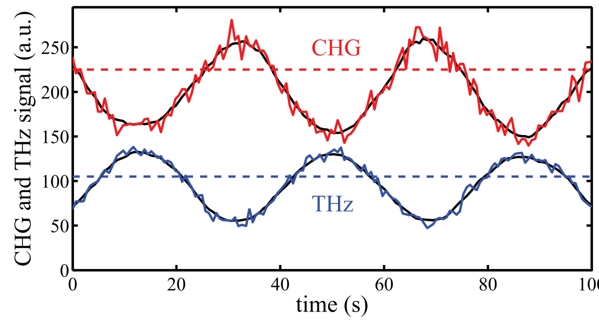

Figure 7: Slow oscillation of CHG and THz intensities due to an RF phase modulation in the ‘breathing regime’ (Fig. 6, center) close to a multiple of the laser repetition rate. The dashed lines indicate the signal intensities without RF phase modulation.

strongly depends on the energy spread, which is large when the bunches are short and vice versa.

For the intended user operation of the short-pulse facility in the presence of an RF phase modulation, a perfect synchronization between the laser pulses and the modulation phase is required. This is easily achieved at DELTA since most time-critical components receive a common 10-MHz signal from an atomic clock.

The CHG intensity and its fluctuations were recorded for different phases of the bunch length oscillation (Fig. 8). At an optimum phase within the ‘breathing regime’ (Fig. 6, center), a CHG intensity of up to 30% higher than without the RF phase modulation was observed indicating that the bunches are even slightly shorter at optimum phase than without modulation. However, the effect of an increased beam lifetime is more pronounced at larger modulation amplitudes (‘island regime’, Fig. 6, right).

Recent studies concentrate on a hybrid filling pattern with a high-current single bunch in the gap of a 3/4 multibunch pattern. Here, the RF phase modulation can be applied to all bunches, while the high-current single bunch is stabilized using a digital bunch-by-bunch feedback system [22]. First promising results show that it is possible to increase the beam lifetime without reducing the CHG intensity.

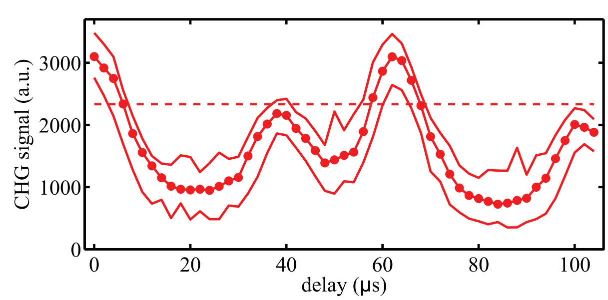

Figure 8: CHG intensity versus delay of the RF modulation phase (‘breathing regime’, Fig. 6, center) showing a minimum, maximum, and average (dots) intensity during a 20 second interval. The dashed line indicates the CHG intensity without RF phase modulation.

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SUMMARY AND OUTLOOK

Since commissioning of the short-pulse facility in 2011, extensive characterization and optimization studies of the CHG and coherent THz radiation have been performed. CHG operation has already been performed during user shifts employing a hybrid filling pattern. Studies using an RF phase modulation have shown that the CHG signal can be significantly increased. Preparations for first user experiments are nearly completed. The final goal of the CHG facility is the generation of ultrashort pulses at 23 eV photon energy (53 nm, fifth harmonic of 266-nm seed), while a first pump-probe experiment will be performed at 9.3 eV photon energy (133 nm, third harmonic of 400-nm seed).

In addition, an upgrade of the short-pulse facility at DELTA using the echo-enabled harmonic generation (EEHG) scheme is in preparation [12].

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