PILOT EXPERIMENTS AND NEW DEVELOPMENTS AT THE DELTA SHORT-PULSE FACILITY*

S. Khan†, B. Büsing, F. Götz, M. Jebramcik, N. M. Lockmann, C. Mai, A. Meyer auf der Heide, R. Niemczyk, B. Riemann, G. Shayeganrad, M. Suski, P. Ungelenk, D. Zimmermann,
Zentrum für Synchrotronstrahlung (DELTA), TU Dortmund, 44227 Dortmund, Germany
M. Plötzing[1], S. Döring[1], M. Gehlmann[1], S. Cramm, L. Plucinski[1], C. M. Schneider[1],
Peter Grünberg Institut, PGI-6, Forschungszentrum Jülich, 52425 Jülich, Germany
[1]also at Fakultät für Physik, Universität Duisburg-Essen, 47057, Germany
S. Xiao, Andrea Eschenlohr, Manuel Ligges, U. Bovensiepen,
Fakultät für Physik, Universität Duisburg-Essen, 47057, Germany

Abstract

At the 1.5-GeV synchrotron light source DELTA operated by the TU Dortmund University, ultrashort radiation pulses in the vacuum ultraviolet (VUV) and terahertz (THz) regime are routinely generated by the interaction of electron bunches with femtosecond laser pulses. A laser-induced energy modulation is converted into a density modulation (microbunching) by a magnetic chicane, leading to coherent emission at harmonics of the initial laser wavelength (coherent harmonic generation, CHG). Path length differences of the energy-modulated electrons along the magnetic lattice lead to a dip in the longitudinal charge distribution, which gives rise to the coherent emission of THz radiation. In first pump-probe photoemission experiments, the spatial and temporal overlap of laser pump and CHG probe pulse on the sample was demonstrated. Furthermore, the effect of two temporally separated seed pulses was studied in the VUV and (sub-)THz regime.

INTRODUCTION

Synchrotron radiation at short wavelengths is the standard tool to study the structure of matter on the atomic scale. However, synchrotron light pulses with durations of 30 to 100 ps (FWHM) are too long to temporally resolve atomic processes taking place on the sub-picosecond scale.

The need for radiation with short wavelength *and* short pulse duration has prompted new developments such as free-electron lasers (FELs) providing extremely brilliant short-wavelength radiation with femtosecond pulse duration. Presently, four linac-based high-gain FEL facilities at sub-visible wavelengths are in user operation and four new facilities are under commissioning. While these FELs are essentially single-user machines (or may serve few users by switching between beamlines), about 50 synchrotron light sources worldwide [1] supply multiple beamlines simultaneously. It is therefore worthwhile to study methods which allow to generate shorter pulses at conventional synchrotron light sources. Some of these methods are adopted from FEL seeding schemes in which the electric field of a femtosecond laser pulse modulates the energy of electrons within a short "slice" at the center of a longer electron bunch. In a scheme known as coherent harmonic generation (CHG, Fig. 1) [2], a magnetic chicane converts the laser-induced energy modulation generated in an undulator ("modulator") into a density modulation (microbunching) giving rise to a short and intense pulse of coherent radiation at harmonics of the laser wavelength in a second undulator ("radiator").

At the 1.5-GeV synchrotron light source DELTA operated by the TU Dortmund University, first attempts were made to perform pump-probe experiments with CHG radiation. Measurements probing the surface state of Cu(111) show a shift and broadening of the photoelectron energy which depends on the pump-probe delay and is attributed to the space charge of the pump-pulse electron cloud. This result clearly demonstrates the spatial and temporal overlap of the two pulses on the sample and constitutes, to the best of our knowledge, the first time-resolved study using photoelectrons produced by CHG pulses.

It is planned to extend the short-pulse facility to shorter wavelengths using the echo-enabled harmonic generation scheme (EEHG) [3] which requires two modulators, two chicanes, and a radiator in a long straight section [4]. Preparatory experiments were performed modulating the electron energy with two laser pulses of equal wavelength, as reported below, and with pulses of different wavelength, see [5].

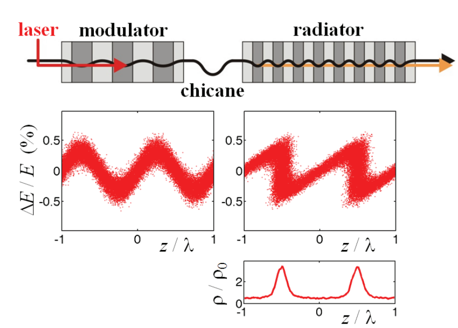

Figure 1: Sketch of the short-pulse scheme CHG with the electron distribution in phase space (relative energy deviation $\Delta E/E$ versus longitudinal coordinate z in units of the laser wavelength λ) and electron density $\rho(z/\lambda)$.

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† shaukat.khan@tu-dortmund.de

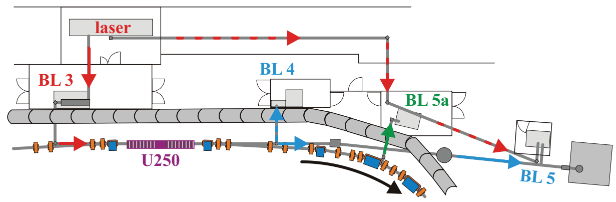

Figure 2: The short-pulse facility at DELTA comprising a laser system, a laser beamline (BL 3) guiding seed pulses to the undulator U250, a diagnostics beamline (BL 4), a soft-x-ray beamline (BL 5), and a THz beamline (BL 5a).

THE SHORT-PULSE FACILITY AT DELTA

In 2011, a short-pulse facility based on CHG was constructed at DELTA to generate ultrashort coherent synchrotron radiation pulses in the vacuum ultraviolet (VUV) and terahertz (THz) regimes [6, 7]. The setup is shown in Fig. 2. Parameters of the storage ring, the laser system, and the undulators are given in Table 1.

Seed pulses from a titanium:sapphire laser system are either focused directly through a beamline (BL 3) into the electromagnetic undulator U250 or are frequency-doubled first. The undulator coils can be powered such that the 7 upstream/downstream periods act as modulator/radiator for CHG with a chicane between them. The energy modulation is performed at the full beam energy of 1.5 GeV.

A diagnostics beamline (BL 4) is used to observe the spatial overlap of laser and undulator radiation on screens and to establish the temporal overlap using a streak camera. In air, spectra down to wavelengths of 190 nm are recorded using a Czerny-Turner spectrometer either by rotating a grating while measuring the radiation intensity with a photodiode or with a fixed grating by using an image-intensified CCD (iCCD) camera [8] with single-shot capability.

Table 1: Parameters of the DELTA Short-Pulse Facility

electron storage ring	
beam energy	1.5 GeV
circumference	115.2 m
beam current (single-/multibunch)	20/130 mA
horizontal emittance	15 nm rad
relative energy spread	0.0007
typ. bunch length (FWHM)	100 ps
titanium:sapphire laser system	
wavelength	800 nm
pulse energy at 800/400 nm	8.0/2.8 mJ
repetition rate	1 kHz
min. pulse duration (FWHM)	40 fs
undulators and chicane	
modulator/radiator period length	250 mm
number of modulator/radiator periods	7
undulator periods used as chicane	3
max. modulator/radiator K parameter	10.5
max. chicane r_{56} value	130 μm

A soft-X-ray beamline (BL 5) operated by the Forschungszentrum Jülich is equipped with a plane-grating monochromator and a hemispherical photoelectron spectrometer with a two-dimensional delay-line detector (DLD) for angle-resolved photoemission spectroscopy (ARPES). In addition to measuring the angular and kinetic energy distribution of valence-band photoelectrons, the timing capability of the DLD was enabled in order to select the CHG-induced electrons. For pump-probe experiments, an evacuated beamline sends a fraction of each laser pulse to the BL 5 endstation. Feedback loops stabilize the position of these pump pulses on the sample. The arrival time relative to the CHG probe pulses is controlled on the 10-fs level by adjusting the optical path length with a motorized linear stage.

The short-pulse facility also comprises a dedicated beamline for terahertz (THz) radiation from a dipole magnet. The energy-dependence of the electron path in the storage-ring lattice causes a sub-picosecond dip in the longitudinal electron distribution which gives rise to coherent emission of THz and sub-THz radiation over several turns [9, 10].

PUMP-PROBE EXPERIMENTS

As a precursor to pump-probe experiments on the sub-ps scale, the Cu(111) surface state was probed by CHG pulses at a wavelength of 133 nm (third harmonic of 400-nm seed pulses) while 800-nm pump pulses were used to create an electron cloud via multiphoton excitation. It is well known that time-resolved photoelectron spectroscopy is prone to spectral shifts by the Coulomb repulsion from the electron clouds induced by both, the pump and the probe pulse [11–13]. These space-charge effects fundamentally limit the applicable pulse energy. On the other hand, a moderate energy shift may be corrected, if properly understood, and it can even beneficial for finding the temporal overlap between pump and probe pulse. The energy shift is detected on the ns scale and peaks at zero delay.

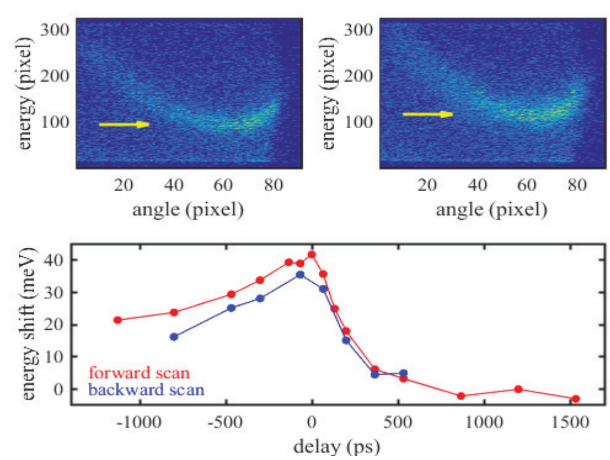

Figure 3: Raw data from the DLD at pump-probe delay 1500 ps (top left) and at 0 ps (top right) with an apparent energy shift of the Cu(111) surface state which depends on the delay (bottom). The difference between the forward and backward scan is consistent with an additional shift depending on the bunch current and thus on the CHG intensity.

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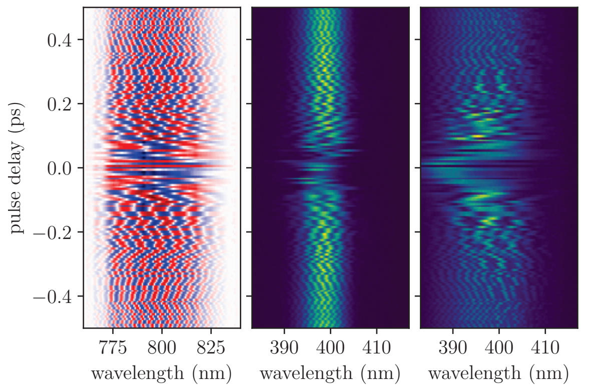

Figure 4: Under variation of the delay between two laser pulses, their spectral interference pattern (left) and interference of 400-nm CHG radiation are shown for two distinctly different cases, a small chirp precompensating the effect of propagation through the vacuum window (center, pulse length 50 fs) and a larger chirp (right, pulse length 90 fs).

In several measurements at DELTA, the space-charge effect was clearly detected demonstrating the spatial and temporal overlap of laser and CHG pulses on the sample. A shift of the photoelectron energy over a delay range >1ns was observed (see Fig. 3) as well as a broadening of the energy distribution over ±100 ps around delay zero. The dependence on delay is a signature of Coulomb repulsion from the pump-pulse electron cloud. In addition, a shift and broadening was identified which did not depend on pump-probe delay but on the CHG pulse intensity indicating a space-charge effect among the probe-pulse electrons.

Simulations were performed tracking a single photoelectron liberated by the probe pulse. For negative delay (pump after probe pulse), this electron is pushed all the way by the pump-pulse electrons. For positive delay (probe after pump pulse), the fast probe-pulse electron travels through the slowly expanding cloud of pump-pulse electrons by which it is first decelerated and then accelerated. Its final kinetic energy depends critically on the spectrum, density and angular characteristics of the pump-pulse electrons. For reasonable parameters, a good qualitative agreement between simulation and measurement is obtained.

SEEDING WITH DOUBLE PULSES

The EEHG scheme requires a twofold energy modulation of the same electrons with two independently focussed laser pulses. As a first step, double pulses were produced by splitting and recombining 800-nm laser pulses in a Mach-Zehnder geometry with one arm comprising a motorized delay stage with a minimum delay step of 200 nm. Sufficient collinearity of both pulses with the electron beam axis was verified by coherent emission of CHG as well as THz radiation with each pulse while blocking the other.

The delay between the pulses was coarsely determined by the respective centroids of CHG and THz intensity as function of pulse arrival time relative to the electron bunch which is controlled by a vector modulator acting on the 500 MHz reference of the laser oscillator. On the femtosecond scale, the relative delay was calculated from the interference pattern of the two pulses recorded using a standard CCD spectrometer, as shown in the left part of Fig. 4. Close to zero delay, interference fringes also show up in the spectra of CHG radiation demonstrating that the phase relationship between the two seed pulses is retained in the microbunching pattern and the coherently emitted radiation. As an example, Fig. 4 shows CHG spectra around 400 nm under variation of the delay between two nearly unchirped (center) and two strongly chirped (right) laser pulses. The dimensionless chicane strength parameter B was set to the rising slope of the second-harmonic bunching factor [3]

$$b_2 = 2e^{-4B^2/2}\,|J_2(2AB)| \tag{1}$$

with A being the energy modulation amplitude normalized to the energy spread. Here, the Bessel function J_2 provides the nonlinearity required in a FROG device [14] for spectrotemporal short-pulse characterization. Thus, CHG with double pulses can act like a collinear interferometric FROG [15].

For a delay in the picosecond range, interference is also observed in THz spectra directly after the laser-electron interaction and in sub-THz spectra one revolution later. The latter regime has been made accessible for spectral studies by a recently commissioned Martin-Puplett spectrometer described in [10]. Figure 5 shows examples of sub-THz spectra for a 0, ±3, and ±6 mm separation (delay 0, ±10, ±20 ps) between two dips in the longitudinal charge distribution.

As described in [5], seeding was also accomplished with two independently focussed pulses at 800 nm and 400 nm, respectively, modulating the electron energy in separate undulators tuned to the respective wavelength.

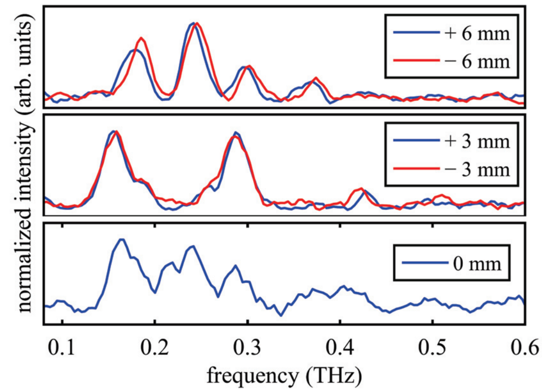

Figure 5: Sub-THz spectra one turn after the interaction between electrons and two laser pulses separated by 0, ±3, and ±6 mm. The symmetry with respect to the sign of the separation confirms the zero-delay assumption.

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