

Spectrotemporal properties of the coherently emitted synchrotron radiation: A machine learning approach

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At the 1.5-GeV synchrotron light source DELTA operated by TU Dortmund University, intense and ultrashort radiation pulses in the vacuum ultraviolet and terahertz regimes are generated by the interaction of femtosecond laser pulses with electron bunches in an undulator. The resulting periodic modulation of the electron energy is converted into a density modulation in a dispersive section, giving rise to coherent radiation emission at harmonics of the seed wavelength in a second undulator. This process is known as coherent harmonic generation (CHG). This paper describes the experimental setup and concentrates on the spectral aspects of CHG radiation under variation of properties of the seed laser pulse and the dispersive section. CHG spectra at the second harmonic of the 800 nm seed wavelength were recorded using a grating spectrometer equipped with an image-intensified CCD camera. Numerical simulations to model the CHG spectra for different group-delay dispersion and third-order dispersion were carried out, and a convolutional neural network was implemented to predict these parameters from measured CHG spectra. The results show that the tunable laser parameters directly translate into the spectrotemporal properties of the coherently emitted pulses.

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I. INTRODUCTION

Modern synchrotron radiation (SR) sources based on electron storage rings are among the primary tools in materials research, physics, chemistry, and biology to study the structure of matter on the atomic scale with short-wavelength radiation [1]. However, phase transitions, chemical reactions, and changes in the electronic or magnetic structure take place on the sub-picosecond scale, which cannot be resolved by conventional synchrotron radiation pulses, which are constrained to tens of picoseconds by the electron bunch length. The femtosecond timescale has been accessed by lasers at near-visible wavelengths, extended by high-harmonic generation to the vacuum ultraviolet range [2]. More recently, high-gain free-electron lasers (FELs) have become available, providing intense femtosecond pulses in the extreme ultraviolet and X-ray regimes [3]. High-gain FELs serve one user at a time at the repetition rate of a linear accelerator (about 100 Hz for normal-conducting and up

to 1 MHz for superconducting resonators), and around ten of these facilities exist worldwide. On the other hand, there are about 50 SR sources supplying multiple beamlines simultaneously with stable and tunable radiation pulses at a rate of up to 500 MHz [4]. It is, therefore, worthwhile to consider possibilities of extending the properties of existing and future SR sources toward shorter pulse durations.

In electron storage rings, a bunch length of a few picoseconds is typically obtained by reducing the momentum-compaction factor [5] or increasing the radiofrequency (rf) voltage. However, pulses with a duration of 100 fs or below can be extracted from a short “slice” within an electron bunch by interacting it with a femtosecond laser pulse in an undulator, which causes a periodic modulation of the electron energy within that slice [6].

Laser-based schemes to generate ultrashort radiation pulses in SR sources are outlined in Sec. II. This paper concentrates on the coherent harmonic generation (CHG) scheme [7] employed at the 1.5-GeV storage ring DELTA [8], which is operated by TU Dortmund University in Germany as a synchrotron radiation user facility and for accelerator physics research. Under variation of the input parameters, CHG spectra show a surprisingly complex structure with interference fringes that are not easily related to the laser properties discussed in Sec. III. After introducing the experimental setup at DELTA in Sec. IV,

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results are shown in Sec. V that were obtained by training a convolutional neural network (CNN) with simulated CHG spectra to obtain the nonlinear dispersion parameters of the laser pulses. Conclusions are offered in Sec. VI, and technical details of the simulation and the neural network design are given in the appendices.

II. LASER-BASED SCHEMES TO GENERATE ULTRASHORT PULSES

Common to most laser-based short-pulse seeding schemes [6] is a periodic energy modulation of electrons co-propagating with a femtosecond laser pulse in an undulator, also called “modulator.” Another possibility, not discussed here, is Compton scattering between laser pulses and an electron beam [9]. In a storage ring, the electron bunches are usually 10^3 times longer than the pulses from a standard femtosecond laser system which defines the above-mentioned slice within which the electron density can be assumed to be constant. Energy modulation is obtained when the electron bunches and laser pulses overlap in all transverse and longitudinal phase space coordinates, i.e., horizontal and vertical position and angle longitudinal position, and when the laser spectrum centered at wavelength λ_L overlaps with the undulator spectrum of fundamental wavelength

$$\lambda = \frac{\lambda_U}{2\gamma^2} \left(1 + \frac{K_U^2}{2} + \gamma^2 \theta^2 \right). \quad (1)$$

This relation implies that the electrons with Lorentz factor γ lag behind the laser phase by one wavelength per undulator period λ_U [10]. The electron velocity along the undulator is controlled by the undulator strength parameter $K_U = 93.4 \cdot \lambda_U[\text{m}] \cdot B_0[\text{T}]$, where B_0 is the magnetic field amplitude. The fundamental wavelength is redshifted at nonzero observation angles θ with respect to the undulator axis. The periodicity of the energy modulation is given by the laser wavelength, and its envelope reflects the shape of the laser pulse. Considering a Gaussian laser pulse with a root-mean-square (rms) transverse size σ_r , the maximum of the sinusoidal energy modulation for an electron located at radius r is given by [11,12],

$$\Delta E(r) = \sqrt{\frac{P_L}{P_0}} \frac{K_U L_U}{\gamma \sigma_r} \left[J_0 \left(\frac{K_U^2}{4 + 2K_U^2} \right) - J_1 \left(\frac{K_U^2}{4 + 2K_U^2} \right) \right] \times m_0 c^2 \exp \left(-\frac{r^2}{4\sigma_r^2} \right), \quad (2)$$

where L_U is the undulator length, J_0 and J_1 are the Bessel functions of the zeroth and first order, m_0 is the electron rest mass, c is the velocity of light, P_L is the peak laser power, and $P_0 = I_A m_0 c^2 / e \approx 8.7$ GW with the electron charge e the Alfvén current $I_A \approx 17$ kA.

Under realistic conditions, the modulation amplitude can be significantly lower than predicted, as it has been observed in several prior studies [13,14], which may be attributed to insufficient laser-electron overlap, optical aberrations, laser wavefront distortions, or other experimental shortcomings.

The energy modulation produced by laser-electron interaction in the modulator can be employed in different ways to generate ultrashort pulses of synchrotron radiation in a second undulator, the “radiator.”

One method, proposed in [15] and often referred to as “femtoslicing,” uses a transverse dispersive displacement of off-energy electrons to separate the short radiation pulse from the long pulse generated by the whole electron bunch. With both radiation components being incoherently emitted, their intensity ratio is typically 10^{-4} . This method was pioneered at the ALS in Berkeley, USA [13], and is currently employed at BESSY in Berlin, Germany [14].

Another way to make use of the laser-induced energy modulation is to convert it into a density modulation by subwavelength path length differences

$$\Delta z = R_{56} \frac{\Delta E}{E_0}, \quad (3)$$

in a dispersive section between the modulator and radiator, which is usually a “chicane” composed of four dipole magnets and characterized by the transfer matrix element R_{56} [10], also known as longitudinal dispersion. In this way, a sequence of microbunches with an equal spacing of the laser wavelength λ_L is created, which gives rise to coherent emission at this wavelength and harmonics thereof [7]. This scheme is called coherent harmonic generation (CHG) and resembles an FEL seeding scheme known as high-gain harmonic generation (HGHG), which was proposed and demonstrated at Brookhaven National Laboratory in Upton, USA [16,17], and is in use, e.g., at FERMI [18], a seeded FEL user facility in Trieste, Italy.

In a storage ring, however, the peak current of the electrons is not sufficient to allow significant FEL gain in the radiator. Here, the goal is to produce a short pulse of coherently emitted radiation, which is more intense than the long incoherent pulse from the whole bunch. CHG was first demonstrated at ACO in Saclay, France [19], and was implemented at several storage rings to generate short radiation pulses, e.g., at UVSOR in Okazaki, Japan [20]; Elettra in Trieste, Italy [21]; and DELTA in Dortmund, Germany [22,23].

The power of radiation emitted in a small interval $d\omega$ around a frequency ω by N electrons is given by the sum of incoherent and coherent emission [10]:

$$\begin{aligned} P(\omega)d\omega &= P_{\text{inc}}(\omega)d\omega + P_{\text{coh}}(\omega)d\omega \\ &= P_e N d\omega + P_e N(N-1)b^2(\omega)d\omega \end{aligned} \quad (4)$$

with

$$b(\omega) = \left| \frac{1}{N} \sum_{k=1}^N e^{-i\omega z_k/c} \right|, \quad (5)$$

where $P_e(\omega)$ is the power emitted by a single electron. The so-called bunching factor $b(\omega)$ is the normalized sum of complex phasors, each depending on the individual electron position z_k . For a large number of randomly distributed electrons, the expectation value of the bunching factor is $b = 1/\sqrt{N}$, while its largest possible value is unity. With $N(N-1) \approx N^2$, the power ratio between the coherent emission from N_s electrons in the energy-modulated slice and the incoherent emission from N_b electrons in the whole bunch is

$$\frac{P_e(\omega) d\omega N_s^2 b^2(\omega)}{P_e(\omega) d\omega N_b} = \frac{N_s^2}{N_b} b^2(\omega). \quad (6)$$

Assuming $N_b \approx 10^{10}$, $N_s \approx 10^7$, and $b_n \approx 0.1$, the ratio would be 10^2 . Even though the short pulse is produced by a tiny fraction of the electrons, the background signal created by the incoherently emitted radiation from the entire bunch is negligible. Therefore, no spatial separation between the long and short components is required.

Expressing the energy modulation amplitude and the longitudinal dispersion as dimensionless quantities,

$$A \equiv \frac{\Delta E}{\sigma_E} \quad \text{and} \quad B \equiv \frac{2\pi\sigma_E R_{56}}{\lambda_L E_0}, \quad (7)$$

with the nominal electron energy E_0 and the rms energy spread σ_E , the bunching factor for the h th harmonic of the seed wavelength can be shown to be [16]

$$|b(\lambda_L/h)| = \exp\left(-\frac{1}{2}h^2 B^2\right) J_h(hAB). \quad (8)$$

The optimum bunching factor is obtained near the first maximum of the respective Bessel function of the first kind J_h , and it decreases with $\exp(-h^2)$.

A more recently proposed seeding scheme is echo-enabled harmonic generation (EEHG) [12,24], involving a two-fold laser-electron interaction and two dispersive sections. With this more complex phase-space manipulation of the electrons, the bunching factor decreases with $h^{-1/3}$, enabling the generation of higher harmonics compared to CHG or HGHG. The EEHG scheme has been demonstrated at linear accelerators [25–28] up to $h \approx 100$, and a first user facility was commissioned [29]. Using EEHG to generate ultrashort radiation pulses has been proposed for several SR sources [22,30–33], and a first prototype was implemented at DELTA [34]. The following

discussion concentrates on CHG but can also be relevant for more complex seeding schemes.

III. SPECTRAL PROPERTIES OF CHG PULSES

For simplicity, CHG radiation is often described by assuming the electron density as well as the seed laser pulse intensity, and thus the energy modulation amplitude, to be constant. While the longitudinal electron density is indeed nearly constant for a 100-fs slice within a 100-ps-long bunch, the intensity of an ideal Gaussian laser pulse follows a normal distribution in all spatial coordinates. Furthermore, the transverse size of the pulse changes along the modulator, usually with the beam waist at its center. If the waist of the seed pulse is significantly larger than the bunch cross section, a Gaussian longitudinal and flat transverse distribution may be a good approximation. Since the energy modulation amplitude is proportional to the electric field of the seed pulse, i.e., the square root of its intensity, the modulation varies along the slice according to a normal distribution with an rms length larger than that of the seed pulse by a factor of $\sqrt{2}$. Additionally, it must be convolved with a rectangular function of length $\lambda L_U/\lambda_U$ to account for the electron slippage of one radiation wavelength λ per undulator period λ_U .

For a given R_{56} value of the chicane, the bunching factor varies along the slice. Figure 1 shows the calculated bunching factor of the second harmonic using Eq. (8) as a function of the longitudinal position and R_{56} for a modulation amplitude of $A = 5$. The optimum R_{56} setting as a function of position is nearly parabolic. The largest bunching factor is obtained by optimizing R_{56} for the central position, while a larger R_{56} causes overbunching in the central part, leading to two or more smaller maxima along the slice. The intensity of the coherently emitted

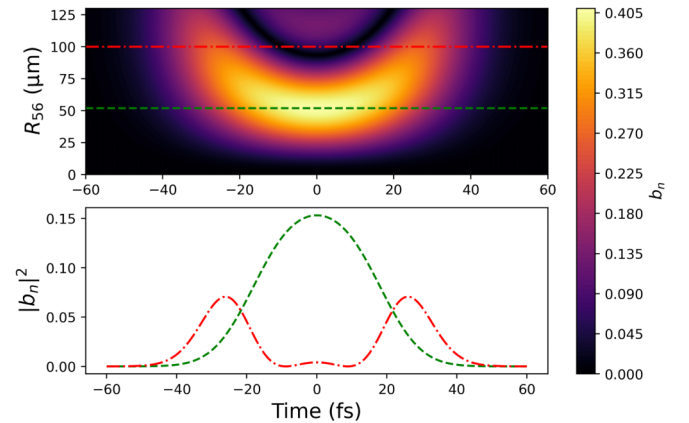


FIG. 1. Top: Calculated bunching factor as a function of longitudinal position (in time units) and R_{56} value of the chicane between modulator and radiator. Bottom: Squared bunching factors for the R_{56} values indicated in the top figure by a red and green line, respectively.

radiation is proportional to the square of the bunching factor, which is shown in the bottom part of Fig. 1 for two R_{56} values. A nearly Gaussian temporal distribution (green dashed line) results in a Gaussian spectrum. Conversely, a temporal distribution consisting of multiple sub-pulses with the same frequency (red dashed line) leads to spectral interference fringes. Neglecting the small central maximum, the resulting spectral shape is analogous to the angular distribution in a double-slit experiment with soft edges. Thus, tuning the R_{56} value enables spectrotemporal shaping of the coherently emitted pulses, which was experimentally demonstrated in the context of HGHG at FERMI in Trieste, Italy [35].

In addition to the chicane strength, the distribution of the spectral phase along the seed laser pulse also influences the spectrotemporal properties of CHG radiation. The electric field of a laser pulse with central frequency ω_0 is expressed in the frequency domain in terms of spectral amplitude $\tilde{\mathcal{E}}(\omega)$ and spectral phase $\varphi(\omega)$ by

$$\tilde{\mathcal{E}}(\omega) = |\tilde{\mathcal{E}}(\omega)| \cdot e^{-i\varphi(\omega)}, \quad (9)$$

where $\varphi(\omega)$ can be expanded into a Taylor series as

$$\begin{aligned} \varphi(\omega) = & D_0 + D_1 \cdot (\omega - \omega_0) + \frac{1}{2} D_2 \cdot (\omega - \omega_0)^2 \\ & + \frac{1}{6} D_3 \cdot (\omega - \omega_0)^3 + \dots \end{aligned} \quad (10)$$

Here, D_0 is the central phase, D_1 is the group delay, D_2 is the group-delay dispersion (GDD), and D_3 is the third-order dispersion (TOD). A transform-limited laser pulse, i.e., a pulse with a minimum time-bandwidth product, is characterized by GDD, TOD, and higher-order terms being zero, while the phase D_0 and a temporal shift due to nonzero D_1 are of no consequence. A nonzero GDD with linear longitudinal variation of the frequency, a linear “chirp,” lengthens the pulse. A nonzero TOD with parabolic frequency variation, in addition, introduces a temporal asymmetry and side pulses preceding or following the main pulse.

When the seed pulse is strongly chirped, two CHG pulses produced at large R_{56} have different frequencies. Figure 2 shows calculated spectra of the CHG radiation for different values of R_{56} , each row being the spectrum for the corresponding chicane strength. This type of plot will be referred to as a spectral map from here on. A comparison of the left and right figures shows the effect of nonzero GDD with TOD and higher-order terms being zero.

The spectral phase properties of seed pulses are given by the laser system and may be further modified by the pulses traversing matter like air, lenses, vacuum windows, etc. A Treacy-type pulse compressor with two gratings [36] in the chirped-pulse amplification scheme [37] of the laser

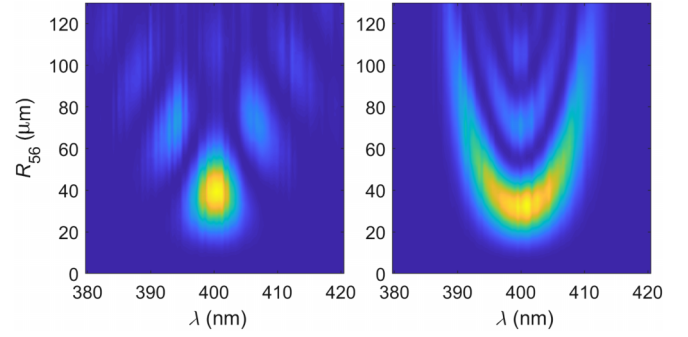


FIG. 2. Calculated spectral maps of CHG radiation, i.e., color-coded intensity (in arbitrary units) as function of wavelength and R_{56} with transform-limited seed pulses (left) and strong chirp (right, GDD = 4000 fs²). All higher-order terms are zero.

system controls the spectral phase properties of the amplified pulse. The GDD and TOD relate to the pulse compressor parameters according to [38]

$$D_2 = -2 \frac{4\pi^2 c d}{\omega_0^3 g^2} \{1 - [G - \sin \alpha]^2\}^{-\frac{3}{2}} \quad (11)$$

$$D_3 = -\frac{D_2}{2\omega_0} \frac{1 + G \sin \alpha - \sin^2 \alpha}{1 - [G - \sin \alpha]^2}, \quad (12)$$

with $G \equiv 2\pi c/(\omega_0 g)$. Here, d is the distance between the two gratings, g is the grating constant, α is the angle of incidence, and c is the speed of light. Tuning d allows changing the GDD and TOD of the compressed laser pulse but does not offer independent control over each.

IV. EXPERIMENTAL SETUP

A. The short-pulse facility at DELTA

DELTA is a synchrotron light source operated by TU Dortmund University for radiation users as well as accelerator physics research. The facility comprises a linear preaccelerator, a synchrotron as a full-energy injector, and a 1.5-GeV storage ring with three insertion devices (a superconducting wiggler, a permanent-magnet undulator, and an electromagnetic undulator). Based on laser-electron interaction in the CHG or EEHG configuration, the short-pulse facility at DELTA produces ultrashort pulses in the vacuum ultraviolet regime as well as coherently emitted terahertz radiation. In the CHG configuration employed until recently, the coils of the electromagnetic undulator U250 were wired such that the first seven periods acted as the modulator, the last seven periods as the radiator, and the three periods in between as a chicane. The modulator-chicane-radiator configuration is also known as an optical klystron [39], reviewed in Ref. [40]. As shown in Fig. 3, the laser pulses are focused into the modulator through beamline BL 3, and the resulting CHG radiation is directed through beamline BL 4 to several diagnostic instruments

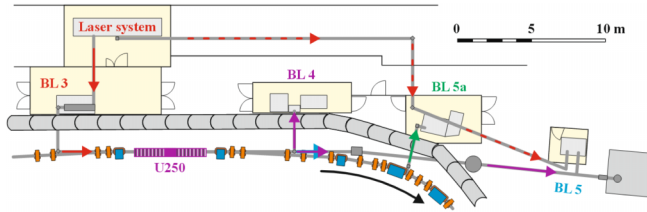


FIG. 3. Short-pulse facility at the DELTA storage ring comprising a laser system, a beamline (BL 3) directing seed pulses into the undulator U250, a diagnostic station (BL 4), a soft x-ray user beamline (BL 5), as well as a THz beamline (BL 5a).

described below. The short-pulse facility also comprises a soft x-ray user beamline (BL 5), a dedicated terahertz beamline (BL 5a), and a laser beamline sending laser pulses directly from the laser system to the experimental stations at BL 5 and BL 5a for pump-probe applications. The parameters of the DELTA storage ring, the femto-second laser system, and the undulator U250 are summarized in Table I.

Figure 4 shows the optical paths of the laser beam and CHG radiation. The laser system comprises a Ti:sapphire oscillator (Coherent Micra) with a repetition rate of 83.3 MHz, the 6th subharmonic of the storage ring rf. The laser amplifier (Coherent Legend Elite Duo HP) consists of a regenerative amplifier, followed by a single-pass stage, both pumped by the same frequency-doubled

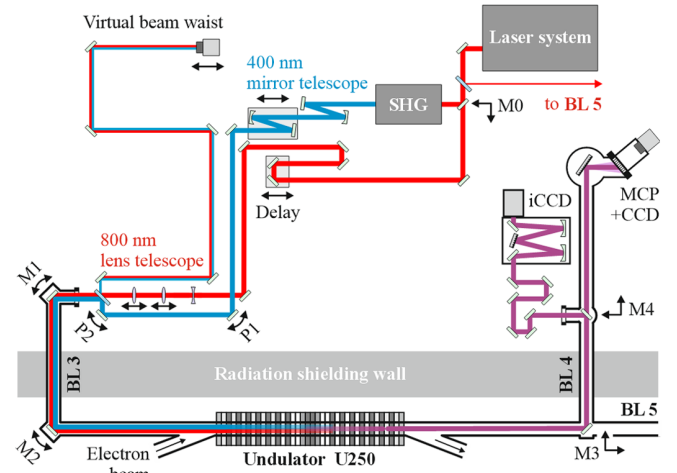


FIG. 4. Schematic setup with laser beam (800-nm red; 400-nm blue) and CHG radiation (magenta). Double arrows indicate movable parts. When mirror M0 is in place, laser pulses from a Ti:sapphire system are frequency-doubled and focused by a mirror telescope. When M0 is retracted, the second-harmonic generation (SHG) unit is bypassed and the 800-nm pulses are focused by lenses. Leakage from a dielectric 400-nm mirror is monitored by a camera (virtual beam waist). Two dielectric 800/400/267-nm mirrors (M1 and M2 at beamline BL 3) align the laser pulses with the electron beam in the undulator U250. CHG pulses are sent by an Al-coated, water-cooled mirror M3 to beamline BL 4 with two alternative grating spectrometers.

TABLE I. Parameters of the DELTA short-pulse facility.

Parameter	Value
Electron storage ring	
Circumference	115.2 m
Revolution frequency	2.603 MHz
Radiofrequency	499.82 MHz
Beam energy	1.5 GeV
Beam energy spread	1.2 MeV
Beam current (multibunch)	130 mA
Beam current (single bunch)	20 mA
Bunch length (FWHM)	30 mm
Electromagnetic undulator U250	
Period length	250 mm
Total number of undulator periods	17
Number of modulator periods	7
Number of radiator periods	7
Maximum modulator/radiator K parameter	10.4
Maximum chicane R_{56} value	170 μm
Titanium:sapphire laser system	
Wavelength	800 nm
Repetition rate	1 kHz
Minimum pulse duration (FWHM)	40 fs
Pulse energy at 800 nm	8.0 mJ
Pulse energy at 400 nm	2.4 mJ

Nd:YLF laser. A pulse energy of 8 mJ with a pulse duration of 40–50 fs is obtained at a repetition rate of 1 kHz, which corresponds to about one laser pulse per 2600 storage ring revolutions. In addition to the 800-nm laser wavelength, seed pulses can be generated at 400 nm by second-harmonic generation (SHG) using a commercial frequency conversion unit (APE TP-1A). The seed wavelength is selected by flipping mirror M0 shown in Fig. 4.

The 800-nm laser pulses are focused by a three-lens telescope. Two of the lenses are motorized to adjust the position and size of the laser waist. For the frequency-doubled pulses at 400 nm, two curved mirrors are employed instead of lenses to avoid excessive group-delay dispersion and intensity-dependent phase shifts. One curved mirror is mounted together with a flat mirror on a linear stage to adjust the laser waist without having to change the mirror angles.

After focusing, leakage from a mirror is transported over a distance equivalent to that of the modulator. At this “virtual beam waist,” a CCD camera on a rail allows inspection of the waist size and shape as well as changes in its longitudinal and transverse positions.

The angles of two in-vacuum mirrors (M1 and M2 in Fig. 4) are remotely controlled to optimize the transverse beam position and angle to maximize the energy modulation of the electrons, which is either judged by the intensity of coherent terahertz emissions measured with a

Schottky diode at the dipole beamline BL 5a or by the CHG signal. In the case of EEHG, two additional piezo-driven mirrors (P1 and P2) allow the alignment of both laser beams simultaneously onto the electron beam axis.

The laser oscillator is synchronized to the storage ring rf, which can be phase shifted using a vector modulator, and one pulse per millisecond is selected for amplification by a set of Pockels cells. In order to synchronize this laser pulse with a single electron bunch in the storage ring, the relative arrival time of the laser and undulator radiation is first coarsely measured using a photodiode at the diagnostics beamline BL 4. Subsequently, a streak camera (Optronis Optoscope) is employed to overlap the pulses with picosecond precision.

With mirror M3 in place, beamline BL 4 is used to characterize CHG radiation with different instruments. For example, the longitudinal and transverse coherence lengths were studied using a Michelson interferometer and a double-slit setup, respectively [41]. The following discussion concentrates on the spectrotemporal properties of CHG radiation.

B. Measurement of CHG spectra

As long as a direct measurement of the temporal CHG pulse shape is beyond reach, only the time-integrated intensity and the spectral properties are experimentally accessible. Spectra of CHG radiation down to 200 nm were obtained using a Czerny–Turner-type monochromator equipped with an image-intensified CCD (iCCD) camera (Andor iStar 334T) installed at BL 4. Gating with a window significantly shorter than the revolution time (384 ns) allows the iCCD camera to record the CHG spectra without the background from the 2600 incoherently emitted pulses between consecutive CHG pulses. For observing radiation below 200 nm, an XUV spectrometer (HP Spectroscopy easyLIGHT) was recently installed at BL 4, which can record spectra from 200 nm down to 30 nm. At the time of the measurements discussed below, a MgF₂ window, which separated the storage ring vacuum from the beamline, blocked wavelengths below 130 nm. Thus, the 6th harmonic of the 800-nm seed was the highest harmonic that could be observed.

Spectral maps of different harmonics of the 800-nm seed were generated by combining the recorded spectra while scanning the current of the chicane magnets from 0 to 700 A in steps of 1 A, corresponding to a R_{56} range of zero to 130 μm . The scan was performed at a rate of 1 A/s with a single spectrum collected at each current step. For the second harmonic, this was repeated for different spectral phases of the seed pulse by changing the separation between the compressor gratings inside the laser amplifier. The GDD and TOD values are dictated by Eqs. (11) and (12), where the parameters of the compressor installed at DELTA are given by $g = 1500^{-1}$ mm and

$\alpha = 51.3^\circ$. The grating distance corresponding to the shortest laser pulse (around 40 fs FWHM) is $d \approx 0.265$ m.

V. EXPERIMENTAL RESULTS AND ANALYSIS

Figure 5 shows the spectral maps of the CHG radiation at the second harmonic of the 800-nm seed. The left plot corresponds to a seed pulse with no frequency chirp, i.e., zero GDD, and exhibits fringes arising from the interference of the consecutive maxima at large R_{56} values. On the other hand, in the case of a large GDD, the right plot of Fig. 5 shows a parabolic feature similar to the simulations in Fig. 2 (right), but with a noticeable asymmetry in the observed spectra. Figure 2 does not take the higher-order spectral phase into account, which is, however, necessary to describe the observed asymmetry.

A. Analysis with convolutional neural networks

The effects of TOD of the seed laser pulse on the spectral map of the CHG radiation were studied using numerical simulations, since independent control of GDD and TOD was not feasible with the present laser amplifier. Energy modulation using a seed pulse with different values of GDD and TOD was simulated, and the spectra were calculated as the square of the bunching factors for the electron distribution after the chicane. It was observed that a large nonzero TOD in the simulation could reproduce the asymmetric features that were present in the measurements. These arise from the fact that a nonzero TOD introduces an asymmetry in the laser pulse shape, which gets imprinted onto the energy modulation and is reflected in the spectral features it produces. To estimate the GDD and TOD of the seed laser pulse from the observed spectral maps, a CNN [42] was designed with *TensorFlow* [43]. The CNN was trained on a dataset of simulated spectral maps for various combinations of GDD and TOD. These maps, in the range of 380 to 420 nm and for chicane strengths corresponding to R_{56} from 0 to 130 μm , were simulated with *MATLAB* for different values and were then converted to images with a size of 64×64 pixels. To prepare the training samples further, Gaussian noise

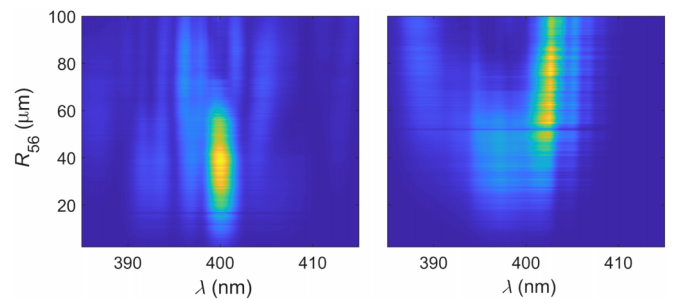


FIG. 5. Observed spectral maps for the second harmonic of the 800-nm seed laser for a pulse with zero (left) and positive frequency chirp (right).

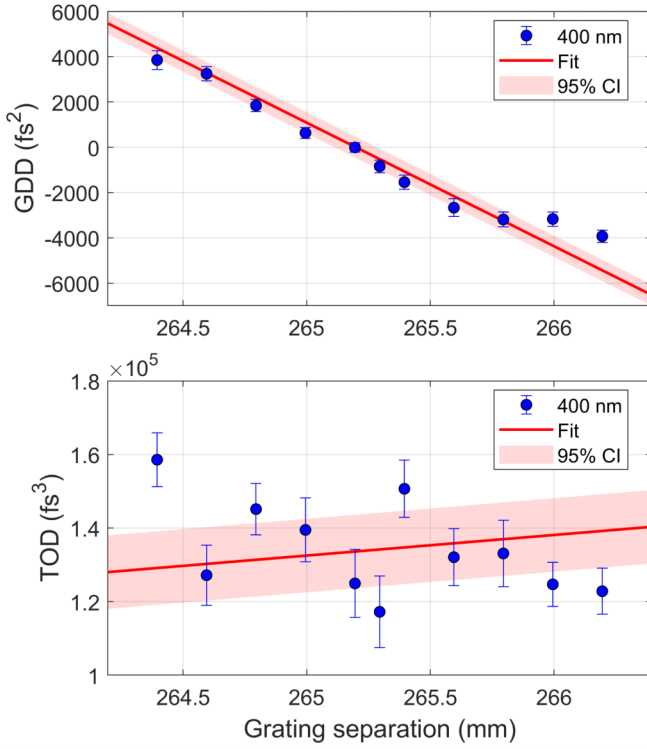


FIG. 6. Blue dots indicate the GDD (top) and TOD (bottom) values predicted by the CNN as a function of the compressor grating separation. Error bars were calculated as the standard deviation of the predictions repeated 100 times. A linear fit to the data and the 95% confidence interval are shown as solid red lines and shaded regions, respectively.

overlays were added, and random translations were performed. The network was trained on a simulated dataset of 43310 samples to predict the GDD and TOD values for which the simulations match the observations most closely. More details are given in the Appendix.

Using the CNN, the GDD and TOD of the seed pulse for 11 different compressor grating settings were predicted. A linear fit by fixing the slopes to theoretical values given by Eqs. ((11) and (12), and using only the offsets as fit parameters was performed as shown in Fig. 6. The shaded regions indicate the 95% confidence interval for the fit. The predictions for GDD were found to be in good agreement with the theory, while those for TOD were associated with larger errors due to the lower sensitivity of spectral features to a change in TOD. The large variance in the TOD values predicted by the CNN may also be the reason for their apparent negative slope, which is not physically meaningful according to Eq. (12).

It should be noted that the TOD is predicted to have large positive values in the order of $1.2 \times 10^5 \text{ fs}^3$, which contradicts the assumption that the stretcher and compressor in the chirped-pulse amplification scheme produce an amplified pulse with zero GDD and TOD. The large positive values of TOD suggest that the TOD introduced by the stretcher may not be fully compensated by the

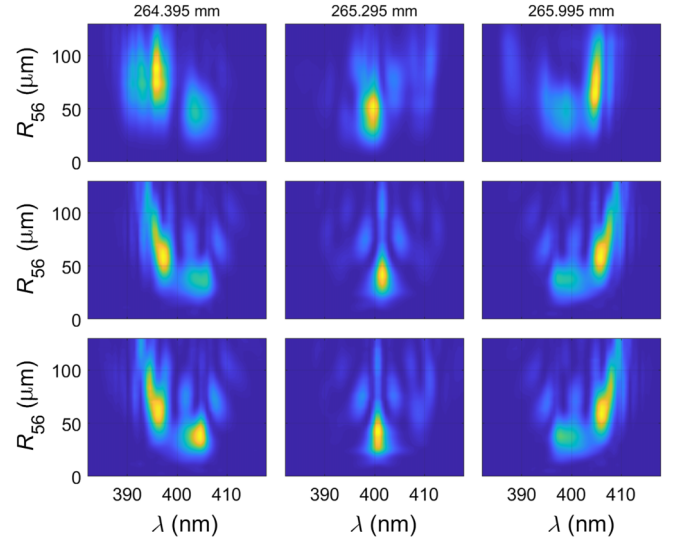


FIG. 7. Observed CHG spectral maps (top row), simulated spectral maps from the GDD and TOD values predicted by CNN (middle row), and spectral maps for GDD and TOD values interpolated from the fit (bottom row) for different compressor grating separations specified above each column.

compressor. The exact source of the TOD remains to be studied.

The predictions were further validated by comparing the measured and predicted spectral maps, examples of which are shown in Fig. 7 for three different separations of the compressor grating pair. With the inclusion of TOD in the simulations, it was possible to replicate the features visible in the recorded spectral maps (top row). The difference between the maps generated from the predicted GDD and TOD values (blue dots in Fig. 6) and the interpolated fit values (on the red lines in Fig. 6) is barely noticeable.

The presence of a significant positive TOD suggests suboptimal conditions for achieving a transform-limited laser pulse, which directly impacts the efficiency and quality of the CHG output pulse. On the other hand, the observed TOD opens a possibility of leveraging it as a tunable parameter for shaping the temporal and spectral characteristics of the CHG emission.

By carefully controlling the TOD, it becomes possible to tailor the pulse shape and spectral content to suit specific experimental needs. For instance, when both TOD and GDD are large, the CHG process in combination with high R_{56} values can generate two temporally separated pulses with distinct central wavelengths and intensities. The pulse shape and spectra for three different combinations of GDD and TOD are depicted in Fig. 8. A zero-TOD condition results in two similar sub-pulses, whereas increasing the TOD allows tuning their asymmetry in intensity as well as pulse length.

For a more quantitative comparison of the features in the observed, predicted, and fitted spectral maps, the positions of the two most prominent peaks visible in the spectra for

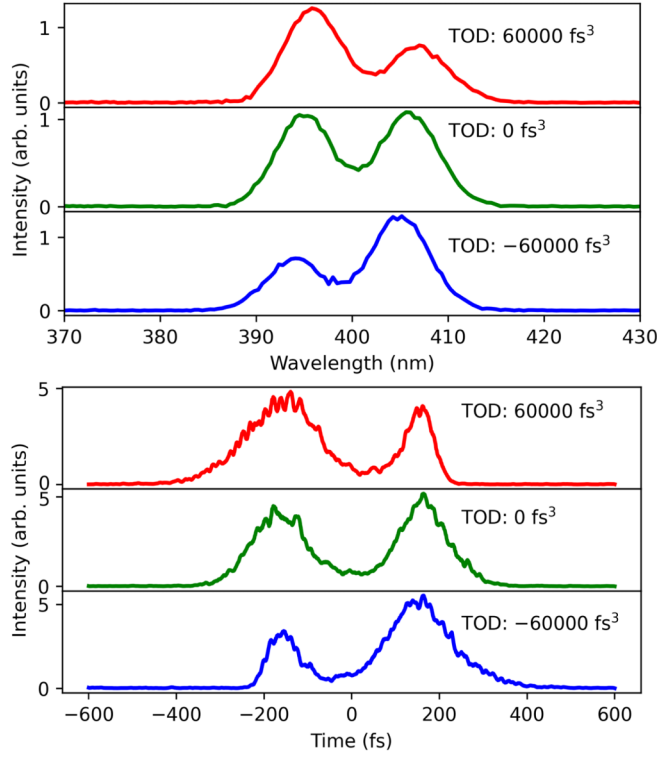


FIG. 8. Spectra of CHG pulses at different TOD values (top) and their respective temporal shape (bottom). All examples are for a GDD of 5000 fs² and an R_{56} value of 50 μ m. The simulations were performed using *SPECTRA* [44].

all 11 samples were calculated. One is the peak around the central wavelength of 400 nm, and the other is a secondary bright peak appearing on one side of the central wavelength. The side on which the secondary peak appears depends on the sign of the GDD. Figure 9 (top) shows the central peak position as a function of grating separation for the observed (red), predicted (blue), and interpolated (green) spectral maps. The central peak position moves from values above 400 nm to values below 400 nm as the grating separation is increased. Similarly, the secondary peak position as a function of grating separation is plotted in Fig. 9 (bottom). Here, all three curves follow the same trend, exhibiting a sharp jump from values below 400 nm to values above 400 nm as the GDD switches from positive to negative. In both comparisons, the observed, predicted, and interpolated values were found to be in good agreement, further validating the efficiency of the CNN.

To quantify the asymmetry in the spectral features, a parameter $\mathcal{A} = |\bar{\lambda} - \hat{\lambda}|$ is introduced as the absolute shift between the spectral peak and the intensity-weighted centroid. The peak wavelength $\hat{\lambda}$ is determined via a smoothing spline fit to the spectrum recorded at the optimal chicane strength R_{56}^* . The centroid $\bar{\lambda}$ is calculated from the projection of the spectral map onto the wavelength axis, $\bar{I}(\lambda)$, as

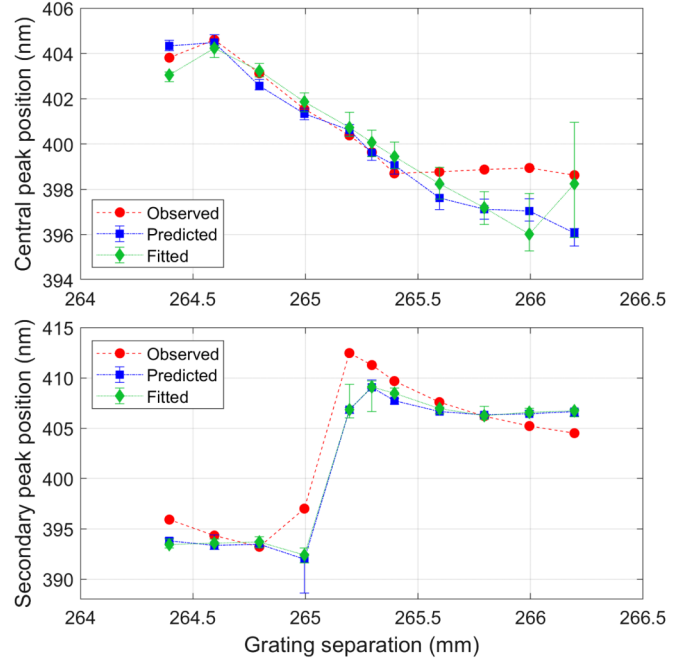


FIG. 9. Central peak position (top) and secondary peak position (bottom) in the observed (red), predicted (blue), and interpolated (green) spectral maps as a function of the grating separation.

$$\bar{\lambda} = \frac{\int \lambda \bar{I}(\lambda) d\lambda}{\int \bar{I}(\lambda) d\lambda}. \quad (13)$$

A value of $\mathcal{A} \approx 0$ indicates a symmetric distribution, while larger values correspond to significant side peaks or spectral lopsidedness induced by GDD and TOD. The asymmetry parameter is plotted in Fig. 10 as a function of grating separation. A dip in the asymmetry around a grating separation of 265 mm was observed, which corresponds to nearly zero GDD. The asymmetry parameters for the observed spectra show good agreement with the values for the predicted and interpolated spectra.

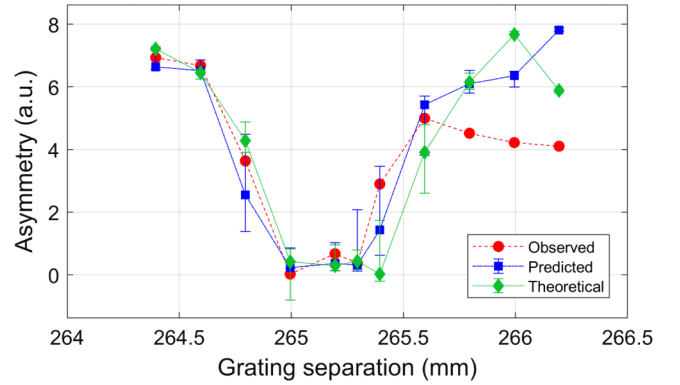


FIG. 10. Asymmetry parameter (see text) as a function of grating separation for observed (red), predicted (blue), and interpolated (green) spectra.

At large GDD values, the central spectral peak becomes broad, and its position is less well defined, as visible in the left and right columns of Fig. 7. This uncertainty contributes to the larger discrepancy between observed and predicted asymmetry $\mathcal{A}$ at grating separations above 265.5 mm.

VI. CONCLUSION AND OUTLOOK

In this study, a CNN-based method was successfully implemented to gain deeper insight into the CHG process by predicting the linear and nonlinear chirp parameters of the seed laser pulse from the spectral distributions. While GDD influences the occurrence of interference fringes at large R_{56} values, the present study indicates that the seed pulses contained a large TOD. The resulting asymmetry in the spectral profile is much larger than that observed in similar studies performed at the seeded free-electron laser FERMI [35]. The effect of TOD on the spectrottemporal properties of coherently emitted pulses in seeding schemes like CHG or HGHG has been largely unexplored. The present results indicate that separate control of GDD and TOD can be a valuable tool for spectrottemporal pulse shaping. Diagnosing the seed laser properties directly from the spectral properties of the coherently emitted radiation pulses demonstrates the predictive power of the CNN-based method.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

APPENDIX A: SIMULATION OF THE CHG SPECTRA

To model laser pulses for different GDD and TOD values, a transform-limited pulse length Δt_0 of 45 fs (FWHM) was assumed. The spectrum was assumed to be a Gaussian centered around a wavelength of 800 nm, corresponding to an angular frequency of $\omega_0 = 2.35 \times 10^{15} \text{ s}^{-1}$. Given by the time-bandwidth product of transform-limited pulses, the angular frequency bandwidth is $\Delta\omega_0 = 6.16 \times 10^{13} \text{ s}^{-1}$

(FWHM). To describe the electric field of a pulse as a function of time, monochromatic waves are summed up as

$$\mathcal{E}(t) = \sum_{\omega=\omega_0-\Delta\omega}^{\omega_0+\Delta\omega} \tilde{\mathcal{E}}(\omega) e^{i(\omega t + \varphi(\omega))}, \quad (\text{A1})$$

where $\tilde{\mathcal{E}}(\omega)$ is the spectral amplitude, and $\varphi(\omega)$ is the spectral phase given by Eq. (10). Here, $\Delta\omega$ is chosen as $5\Delta\omega_0$ such that the frequency range is considerably larger than the bandwidth of the pulse. The electric field is then normalized to its maximum

$$\hat{\mathcal{E}}(t) = \frac{\mathcal{E}(t)}{\max\{\mathcal{E}(t)\}}. \quad (\text{A2})$$

To create the electron distribution after laser-electron interaction, an initial distribution of 2.4×10^5 particles in the bunch slice (corresponding to 100 nA of beam current in the DELTA storage ring) with an energy spread of 7×10^{-4} was generated. The energy modulation was then applied to the electrons as

$$E_k^f(t_k) = E_k^i(t_k) + \hat{\mathcal{E}}(t_k) \Delta E, \quad (\text{A3})$$

where $E_k^i(t_k)$ and $E_k^f(t_k)$ correspond to the initial and final energy of the k th electron, respectively; ΔE is the modulation amplitude; and t_k is the averaged temporal position of the electron with respect to the laser pulse, here taken at the center of the modulator.

To simulate the transfer through a dispersive chicane, an energy-dependent temporal shift was applied to the electrons as

$$t'_k = t_k + R_{56} E_k^f(t_k), \quad (\text{A4})$$

converting the energy modulation to a density modulation. The spectra of the CHG radiation were calculated as the square of the bunching factor $b(\omega)$ given by

$$b(\omega) = \left| \frac{1}{N} \sum_{k=1}^N e^{-i\omega t'_k} \right|. \quad (\text{A5})$$

To generate the spectral maps of CHG radiation, $b^2(\omega)$ was evaluated for a wavelength range of 380 to 420 nm ($\omega = 4.96 \times 10^{15} \text{ s}^{-1}$ to $4.48 \times 10^{15} \text{ s}^{-1}$) and for R_{56} ranging from 0 to 130 μm .

APPENDIX B: CNNs

Conventional two-dimensional fitting algorithms are limited in their usefulness for comparing observed CHG spectra under variations of the R_{56} value to simulated spectral maps due to the noise contributions and imperfections in the measurements. A better solution is to use a

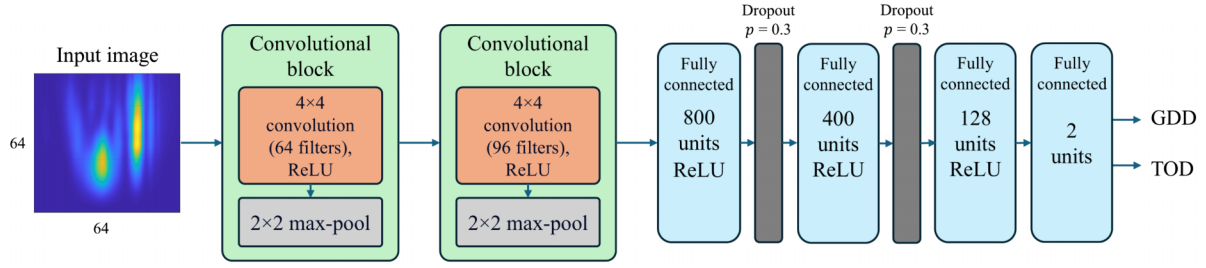


FIG. 11. CNN architecture for predicting GDD and TOD from CHG spectral maps. The model employs two convolutional blocks with max pooling for feature extraction, followed by a series of fully connected and dropout layers to output the GDD and TOD values.

CNN, as the features present in the spectral maps are of greater interest than their absolute positions. In this work, a CNN consisting of two convolutional blocks and three fully connected layers was employed (see Fig. 11). The network takes the 64×64 pixel images as input and has two units in the output layer that give the values of GDD and TOD. The convolution blocks consist of a convolutional layer, followed by a ReLU activation function and a max-pooling layer [45]. The convolutional layers use a 4×4 kernel with 1×1 stride and zero-padding, which applies a set of filters to the input spectral images, effectively acting as feature detectors. The max-pooling layers use a 2×2 kernel with zero padding, which reduces the image size by keeping only the strongest features, making the network robust against small shifts or noise in the measured spectra. The three fully connected layers have 800, 400, and 128 units, respectively. In this study, the Huber loss function [46] and the Adam optimizer [47] with a learning rate of 10^{-4} were used to train and optimize the CNN. The Huber loss function is used here instead of mean squared error because it is less sensitive to outliers.

For error estimation of the predictions, the Monte Carlo dropout technique [48] was adopted. This involves randomly deactivating units in each layer with a given probability ($p = 0.3$) and performing multiple predictions with a different unit being deactivated each time. This results in a distribution of predictions since the model varies slightly with each iteration. The statistics of this distribution can be inferred to estimate the mean and rms error of the predictions.

APPENDIX C: PREPARATION OF THE TRAINING SAMPLES

Since there were no labeled measurements of the CHG spectra, simulated CHG spectral maps served as the training samples for the CNN. CHG spectral maps for different combinations of GDD and TOD were calculated with the GDD ranging from -7000 fs^2 to 7000 fs^2 and the TOD ranging from 0 fs^3 to $1.8 \times 10^5 \text{ fs}^3$. Negative TOD values were excluded from the training samples since the resulting spectral maps are indistinguishable upon inverting the signs of GDD and TOD. It was inferred that the TOD should be

positive by roughly comparing the observed and simulated spectral maps, and, accordingly, only positive TOD values were included in the training samples.

Subsequently, the spectral maps were normalized to their peak intensity and downsampled to images of size 64×64 pixels. To emulate experimental noise, Gaussian noise with an amplitude of 5% was added to the images. The noise does not mask the relevant spectral patterns because it is spatially uncorrelated and the TOD-induced features are structured and distinct in character, the noise does not mask the relevant spectral patterns, as is evident from Figs. 9 and 10. To introduce some variations into the training samples, the images were randomly translated by a few pixels along both axes. The resulting dataset, comprising 43,310 samples, was randomly divided into a training set (80%) and a test set (20%). The training set was used to minimize the loss function, while the test set was used to monitor the model performance on unseen data, verifying the generalization capability of the network and preventing overfitting.

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