Toward AI-enhanced online-characterization and shaping of ultrashort X-ray free-electron laser pulses

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ABSTRACT

X-ray free-electron lasers (XFELs) as the world’s brightest light sources provide ultrashort X-ray pulses with a duration typically in the order of femtoseconds. Recently, they have approached and entered the attosecond regime, which holds new promises for single-molecule imaging and studying nonlinear and ultrafast phenomena such as localized electron dynamics. The technological evolution of XFELs toward well-controllable light sources for precise metrology of ultrafast processes has been, however, hampered by the diagnostic capabilities for characterizing X-ray pulses at the attosecond frontier. In this regard, the spectroscopic technique of photoelectron angular streaking has successfully proven how to non-destructively retrieve the exact time–energy structure of XFEL pulses on a single-shot basis. By using artificial intelligence techniques, in particular convolutional neural networks, we here show how this technique can be leveraged from its proof-of-principle stage toward routine diagnostics even at high-repetition-rate XFELs, thus enhancing and refining their scientific accessibility in all related disciplines.

Introduction

Short-wavelength free-electron lasers (XFELs) are the world’s fastest X-ray cameras, providing ultrashort exposure times in combination with a spatial resolution limit down to the sub-nanometer range, which allows for time-resolved experiments ‘freezing’ the motion of atoms and molecules. In fact, XFELs have revolutionized several fields of science enabling us to observe the role of transient structures and resonances in atoms as well as single-molecule or cluster imaging, investigations of ultrafast processes at element-specific observer sites, and the study of nonlinear light-matter interaction in the X-ray regime.

Over the past decade, further development of the underlying machine operation techniques has enabled increasingly sophisticated control over the photon-pulse parameters. One of the most recent major upgrades is the increased repetition rate of XFELs that is anticipated to initiate a leap from proof-of-principle experiments to advanced applications of interdisciplinary importance, thus representing a cornerstone of modern XFEL science.

Most of the XFELs and in fact all XFELs worldwide are currently based on the principle of self-amplification of spontaneous emission (SASE). More precisely, their pulses are formed stochastically through the interplay between emitted synchrotron radiation caused by sinusoidally undulating electrons and their subsequent density modulation, resulting in ultrashort X-ray pulses of a peak brightness up to and exceeding \(10^{32}\) photons sec\(^{-1}\) mrad\(^{-2}\) mm\(^{-2}\) 0.1%BW (with bandwidth BW). This amplification process generates a non-predictable time–energy structure for every single pulse, constituting one of the biggest limitations for XFEL science so far. There are currently no control mechanisms and no routinely available diagnostics in place to directly measure the temporal properties of these X-ray pulses. Hence, the bulk of dynamics on attosecond to femtosecond time scales occurring during the exposure to the X-rays is unfortunately only inferred via indirect pulse-characterization measurements such as spectral analysis or electron beam diagnostics.
Recently, we have demonstrated a new technique termed angular streaking that is capable of retrieving the time–energy structure of all incoming SASE X-ray pulses non-destructively with attosecond resolution. Besides the major diagnostic breakthrough, this generally paves the way for time-resolved and nonlinear attoscience in the X-ray regime. In fact, the onset of all structural dynamics in matter can now be studied in detail even from specific observer sites through strongly localized electrons. Fast and reliable feedback of this novel diagnostic regarding the experiment and the machine itself is of utmost importance for upcoming scientific applications at XFELs. For high-repetition-rate XFELs such as the European XFEL near Hamburg, Germany, conventional analysis approaches fail to accommodate the enormous amount and complexity of angular-streaking data in full depth. Especially for online analysis and, ultimately, (re)active control and pulse shaping during beam times, conventional data processing methods are not suited. Therefore, several core challenges of XFELs are anticipated to be tackled by machine-learning techniques.

In this article, we present a machine-learning-based proof-of-concept on retrieving the full and detailed XFEL pulse temporal profile, including the pulse duration and its intensity substructure. In addition, we show that it is possible to extract temporal information on the electronic processes after photoionization initiated by the X-rays and a subsequent Auger decay via the method of angular streaking paired with analysis through neural networks (NN). Moreover, by using simulated streaking data with various degrees of instrument noise and different electron emission signatures, we demonstrate the flexibility of the NN-based online diagnostic tool for XFELs. It is thus robust against detector noise and machine fluctuations, and covering the vast majority of current and future operation modes.

Application Case: Angular Streaking

A long-standing goal in laser and X-ray research is to enable measurements providing both temporal and spatial real-time information about structural changes on a molecular level with element-site specificity—a so-called molecular movie. For this, a suitable ultrashort X-ray pulse duration is one of the key parameters, which is both hard to facilitate and difficult to measure. Yet, a reliable time-resolving experimental method is essential for determining parameters such as the detailed intensity profile of the SASE FEL pulse, corresponding damaging thresholds for materials under investigation, the nanoscale interpretation of ultrafast single-shot diffraction imaging, and the probabilities for multi-photon processes, to name a few.

Applying the angular-streaking technique to the field of XFELs leverages a versatile approach for the temporal and spectral characterization of individual (X)FEL pulses. The applied scientific instruments for this new method are angle-resolving electron spectrometers. In case of the first demonstration of angular streaking at XFELs and also in the present case, 16 individually working time-of-flight (TOF) spectrometers are arranged in a ring-like structure around the target region, perpendicular to the propagation direction of the incoming X-rays. Together with a co-propagating circularly polarized infrared laser, spatially and temporally overlapped with the XFEL at the target region, this setup enables angular streaking. Atoms from a target gas are ionized with the XFEL pulse and the emitted electrons are swept, i.e., streaked, in energy and angle by the concomitant rotating electric field vector.
of the infrared laser. In a simplified picture, the streaking field vector can be understood as the hand of a clock that encodes the parameter time via the angles at which electrons are detected with accordingly shifted energies (see illustration in Fig. 2). Given sufficiently many electrons to "report" on their ionization time within the SASE pulse, the measured electron emission patterns contain the information of the full time–energy structure of the ionizing XFEL pulse with attosecond resolution. The method can be adapted for pulses with different photon energy by selecting target gases of suitable electron binding energies and photoionization cross sections. The mechanism and experimental setup for the angular-streaking technique as applied to SASE X-ray pulses is described in [9] and the general principle can be found in [18–20].

Figure 2. Illustration of this work’s topical orientation. A series of previously unknown XFEL pulses releases electrons (indicated in yellow) from a gas target with characteristic kinetic energies. Their kinetic energy shifts caused by a circularly polarized infrared streaking field are recorded in an angle resolving spectrometer (indicated by the cones and eyes). In the artificial brain, the thus obtained information about the electrons is processed and reveals the underlying attosecond substructure of the X-ray pulses. The precise knowledge of the time–energy substructure can then be used to shape the X-ray pulses via a feedback loop to the XFEL machine settings or to enable access to ultrafast electron dynamics at the attosecond frontier, as here indicated by a molecular movie of an interatomic Auger decay in water with subsequent dissociation.

In the experiment under consideration, a time trace is measured for photoelectrons emitted by each X-ray shot and in each TOF spectrometer, hence, generating 16 traces at a rate set by the repetition frequency of the XFEL and of the overlapped streaking laser. For single-shot spectroscopy, a trace represents the number of electrons arriving after specific flight times. These time-domain traces can be converted to the energy domain (spectra) by taking into account the length of the flight path and the actions of additional electric fields along their paths, which are routinely used for enhancing the achievable energy resolution. The combined representation of a full angle-resolved streaking measurement forms an image with 16 columns, representing the respective detector angles, and several rows corresponding to the range of electron energies detected in the specific measurement (detector image, cf. Fig. 1). Time-dependent electron spectra (spectrograms) are then generated by converting the emission angles to times using the known rotation period of the electric field vector for the circularly polarized streaking laser. (cf. Figs. 11a & 11c).

For an X-ray pulse with no temporal and spatial overlap in the interaction region with an external streaking field (unstreaked shot), the spectra in all detectors are showing the characteristic distribution of electron time-of-flights traces (spectral lines) for the target under investigation. Typically each line shows an angular distribution, described in the dipole approximation by the
angular distribution anisotropy parameter $\beta_2$. $\beta_2 = 0$ corresponds to completely isotropic emission, giving rise to the identical signal intensity in each TOF. $\beta_2 = 2$ specifies a $\cos^2$-like emission pattern in the form of a dumbbell oriented in the direction of the (linear) X-ray polarization. In Fig. 1 one can see this $\beta_2 = 2$ distribution in the low intensity regions around $0^\circ$ and $180^\circ$ in contrast to the high-intensity parts at $90^\circ$ and $270^\circ$, with intermediate intensities in the columns at angles in between.

If a circularly polarized streaking laser is present, the detector image is modulated according to the instantaneous streaking laser vector potential, leading to a sinusoidal variation of the spectral lines along the angle axis. The following parameters of the streaking laser are important for this investigation and are kept constant throughout the experiment:

The ellipticity of the polarization is defined as the ratio of the major axes of the polarization ellipse $b/a$. For perfectly circular polarization, this ratio amounts to unity, while it is equal to zero for linear polarization. A small deviation from circular polarization gives rise to a certain degree of ellipticity, which manifests itself in a different maximum streaking amplitude of the electrons for the individual detectors. For an elliptically polarized infrared beam, the principal axis may also have an arbitrary rotation (elliptical tilt) with respect to the ring of detectors, which needs to be reconstructed from the data for an unambiguous X-ray pulse retrieval.

The goal regarding SASE FEL X-ray pulse characterization and their potential control is to reconstruct the spectrogram from a measured detector image, which gives the full information about the X-ray time–energy structure (cf. Fig. 3, dotted line). In many experimental situations, however, it is sufficient to restrict our analysis to some of the most relevant SASE X-ray parameters (cf. Fig. 3, red line). In the subsequent discussion, we have, therefore, focused on the prediction of the temporal aspects of ultrashort FEL pulses. We picked the following pulse characteristics for a comparison of their reconstruction by the NNs (prediction) with the originally simulated data (target):

- **Detector Image**
- **Spectrogram**
- **Relevant Parameters**

**Figure 3.** There are two ways to extract important pulse characteristics from the detector image. The dotted line refers to a full reconstruction (spectrogram) and the extraction of relevant parameters out of this reconstruction. The other way is to skip the full reconstruction and to concentrate on typically most relevant parameters only. In this article, we choose the second approach.

Details about the NN’s framework conditions, the chosen architecture, and hyperparameter optimization can be found in the Methods below.

**Kick** The kick is the maximum streaking shift in electron kinetic energies for each X-ray shot, and thus for a given temporal delay and phase relation between the X-ray pulse and the infrared streaking laser. There are two main reasons for a change in the kick from shot to shot. The first is the relative timing jitter between the X-ray and the streaking pulse, which is unavoidable due to the stochastic generation process of the SASE mechanism and additional fluctuations in arrival time caused by air fluctuations, thermal expansion in optomechanical components, and general synchronization errors between the two separate laser pulses. The second reason for variations of the kick is the random change of the carrier–envelope phase of the streaking laser from shot to shot. One can solve this by stabilizing the carrier–envelope phase, which is a rather difficult technical requirement, or by using the technique of angular streaking, which is the basis for the simulations studied in this article.

**Pulse Duration** The pulse duration is the most important parameter for many ultrafast free-electron laser experiments, e.g., a variety of pump/probe measurements of electronic state changes or investigations of nonlinear excitation dynamics, albeit it is one of the most difficult to measure directly. Especially for XFEL SASE pulses, each pulse has a different duration and erratic intensity structure that even complicates the definition of the term pulse duration. In this article, we use the root-mean-square (RMS) duration, i.e., the square root of the time variance of the temporal intensity profile,

$$t_{p,\text{RMS}} = \sqrt{\langle t^2 \rangle - \langle t \rangle^2},$$

where

$$\langle t^n \rangle = \frac{1}{N} \int_{-\infty}^{\infty} t^n I(t) \, dt, \quad N = \int_{-\infty}^{\infty} I(t) \, dt$$
Figure 4. Three examples of different SASE XFEL pulse intensity structures showing varying total duration and complexity. The FWHM duration for the exemplary shots are 1.46 fs (blue), 4.53 fs (orange) und 10.42 fs (green).

are the $n$-th moment and the normalization constant, respectively, as the definition of the pulse duration.

A common choice for more well-behaved Gaussian-like laser pulses from table-top systems is the full width at half-maximum (FWHM). Given a normal distribution with standard deviation $\sigma$, corresponding to the RMS duration in this case, the FWHM is calculated as follows:

$$FWHM = 2\sqrt{2\ln 2}\sigma \approx 2.35 \cdot \sigma.$$  \hfill (3)

As SASE pulses are generally spiky and irregular (cf. Fig. 4), this metric is not fully applicable. In our simulation case, however, this quantity is nevertheless of interest due to the fact that we use (and in fact know) the FWHM to generate the 2D-gaussian distributions in Fig. 4. The RMS duration, however, gives a more complete measure of the temporal distribution of the pulse energy including possible pulse wings or substructure (see also the next paragraph).

**Pulse Structure** Due to the microbunching in the FEL each SASE pulse has an individual intensity profile, made up of several shorter ‘spikes’ with random intensity (cf. Fig. 4). The average number of spikes per pulse is determined by the specific operation parameters of the XFEL. It can be expressed in a statistical treatment as the number of individual energy modes contributing to the XFEL pulse. The ensuing pulse shape can be arbitrarily complex. The shorter the overall pulse duration in relation to the single-spike length, i.e., the fewer spikes per complete pulse, the more important individual spikes are becoming (Fig. 4). Especially for estimating the damage thresholds of investigated probes as well as for experiments sensitive to the instantaneous X-ray intensity, or for ultrafast pump/probe measurements, the XFEL pulse structure needs to be known exactly to interpret the observed data on a shot-to-shot basis unambiguously.

**Auger Decay Time** Many of the scientifically interesting processes of non-equilibrium physics and structure-changing chemistry are not directly triggered by the exciting X-ray pulse but are the result of subsequent complex relaxation dynamics. These dynamics are determined by the time-dependent, i.e., transient, electronic structure of the system under study. One of the most fundamental electronic processes after inner-shell ionization of matter by X-rays is the Auger decay, whereby a second electron from an outer shell fills the generated core hole and transfers the excess energy to a third electron (Auger electron), which is then emitted from the ion. This process is specific to the contributing discrete electronic states of an atomic or molecular system and has a characteristic time constant for the emission of the third electron (Auger decay time). In our
simulations, we assume that one Auger decay channel dominates for neon (Ne) after 1s ionization. The corresponding Auger decay time on the order of 2 fs to 3 fs\cite{30} can serve as a fundamental benchmark for demonstrating the capability of the method to retrieve ultrafast timing information from recorded data.

Results

All of the above described SASE XFEL pulse characteristics can be predicted with varying degrees of accuracy by utilizing convolutional NN. For each pulse characteristic, we will examine the results of the trained models in more detail.

Kick

Of all the characteristics studied, the kick turned out to be the easiest to predict. Fig. 5a shows that most of the predictions only slightly deviate from the respective targets. When looking at the difference between target and prediction, most of the values do not exceed the limits of 0.5 and −0.5 eV. Though the kick can easily be derived from the detector images, an accurate estimate of this parameter is necessary for better judging the estimates of the characteristics FWHM pulse duration and Auger decay time. This will become apparent in the following paragraphs.

FWHM Pulse Duration

The comparison of predicted and target FWHM pulse durations is shown in Fig. 5b. As for the kick estimates, the majority of the values are well estimated. However, it is evident that some of the values deviate strongly from the target values. One hypothesis explaining this behavior is that for smaller kicks predicting a pulse duration can become arbitrarily difficult. That is why we have investigated the accuracy of the FWHM pulse duration estimate against the true kick value.

Fig. 5c confirms the previously stated hypothesis. Above a (true) kick value of approximately 5 eV, estimating the FWHM pulse duration becomes feasible. This is due to the fact that small kick values correlate to unsuccessful angular streaking shots that need to be discarded anyway. Fig. 6a and 6b display exemplary shots with a small and a large kick, respectively.

Auger Decay Time

The auger decay time can also be well approximated by the respective NN (cf. Fig. 5d). Most of the estimates hardly deviate from the zero line of the difference between the target value and the prediction. However, as for the FWHM pulse duration estimates, there are some outliers strongly deviating from the true Auger decay time value. Using the same reasoning as for the FWHM pulse duration estimates, we have compared the prediction of the Auger decay time value with the true kick value. Here, the same behavior can be observed as for the FWHM pulse duration (cf. Fig. 5e). It is evident that a reasonable determination of the Auger decay time is only possible for a kick value of 3 eV or higher. It follows that the decay cannot be predicted on a failed angular streaking shot either. Shots with small kick values should be discarded in advance to appropriately approximate the true Auger decay time. Fig. 6c and 6d display exemplary shots with a small and a large kick for the decay reconstruction, respectively.

Pulse Structure and RMS Pulse Duration

The full temporal pulse structure of the SASE pulse is probably the most difficult property to predict in our case study. This is not surprising, as it is also the most complex one, being represented by a vector which holds the information of the intensity at equally distributed time steps, mapped to a window of 35 fs later on, as this is the duration of one optical cycle for the chosen streaking wavelength $\lambda = 10.6 \ \mu m$ in Hartmann et al.\cite{9}. We normalize the training data of the time distributions as for the previous labels and, thus, simplify the task for the NN. Altogether, the trained network works relatively well in its objective to predict the trend, i.e., peak positions and their relative intensities, of the pulse structure. However, as has been expected, these predictions get less accurate for more complex pulse structures. This behavior can be seen for two different exemplary simulated SASE pulses in Fig. 8, one relatively simple (a) and one more complex (b), in which not all of the finer structures could be reliably reproduced. At the very least, the main features including the larger peaks can always be predicted.

In contrast to the Auger decay time and the FWHM pulse duration, the quality of the predicted pulse structure does not show a significant dependency on the value of the kick; except for a kick very close to or equalling zero, which is not surprising, as this corresponds to an unsuccessful event where basically no streaking occurred. There is also no significant dependency on the duration of the pulse, as one might have expected. An absolute value for the mean squared error (MSE) does indeed increase with the pulse duration; normalized to it, however, the average ‘MSE per time step’ is more or less constant (cf. Fig. 9a). Now that our model is capable of extracting the pulse structure, it is possible to compute the RMS pulse duration by using Eq. 1. Fig. 9b shows the deviation of this computed RMS pulse duration to the RMS pulse duration of the target pulse structure. The average deviation lies below 1 fs. Only at very long pulses there is a trend towards a slight underestimation, however, the error is still just around 10% in most cases. By computing the full pulse structure, therefore, an additional NN for the prediction of the RMS pulse duration could serve as a comparative value for a coarse estimation of the quality of the reconstructed pulse structure.
(a) Difference between the target and the predicted kick label, demonstrated on a test set with $4 \cdot 10^4$ samples. Most of the values predict the kick very accurately.

(b) Difference between the target and the predicted FWHM pulse duration label, demonstrated on a test set with $4 \cdot 10^4$ samples. There are several outliers that cannot be predicted by the utilized NN.

(c) Comparison of the FWHM pulse duration prediction as a function of kick energy. The accuracy of the pulse length estimate depends on the respective kick value of the shot. The higher the kick, the more accurate the prediction of the FWHM pulse duration gets.

(d) Difference between the target and the predicted Auger decay time, demonstrated on a test set with $4 \cdot 10^4$ samples. Although most predictions are sufficient, there are significant deviations from the target value.

(e) Comparison of the decay prediction as a function of kick energy. The accuracy of the Auger decay time estimate depends on the respective kick value of the shot. The higher the kick, the more accurate the prediction of the Auger decay time gets.

**Figure 5.** Prediction accuracies and dependencies of the labels *kick*, *FWHM pulse duration*, and *decay* using data sets containing all levels of noise [$\pm 0\%$, $\pm 10\%$, $\pm 20\%$, $\pm 30\%$].
### Influence of Noise on the NN Performance

In order to investigate the influence of noise on the predictions, we generated a test set comprising of detector images with several different noise levels \( p = [0.0, 0.1, 0.2, 0.3] \) as shown in Eq. 4 and fed it into the NN. As expected, additional noise influences the result of the NN’s prediction (cf. Tab. 1). Some expressive examples are shown in Fig. 7. The prediction for non-noisy data is nearly perfect, whereas the prediction for noisy data slightly differs from the target. The predicted pulse structures show no real trend for the average MSE’s in dependency of the noise levels, with no significant decrease in quality of the predicted shapes. Despite the decreased prediction accuracies, it is evident that the NNs can handle noise robustly.

### Discussion & Outlook: Online SASE-Pulse Characterization

So far, we have shown that several characteristics of XFEL pulses are predictable with varying degrees of accuracy. To investigate how close the current status comes to real-time pulse characterization during experimental campaigns, we need to...
Figure 8. Examples for simple (a) and more complex (b) simulated and reconstructed SASE pulse structures with RMS pulse durations of 6.20 fs and 11.65 fs, respectively.

Figure 9. Computing the RMS pulse duration from the pulse structure.

Table 1. Standard deviations for the predicted labels kick, simulated pulse duration, and decay computed on 1000 samples, respectively. The overlap between target and predicted pulse structure is shown using the average normalized MSE for several noise levels. The test set used to compute the deviations consists of about $4.6 \cdot 10^4$ samples.

| Noise | Kick  | FWHM  | RMS   | Decay | Structure |
|-------|-------|-------|-------|-------|-----------|
| ±0%   | 0.19 eV | 0.52 fs | 0.67 fs | 0.08 fs | 0.00273   |
| ±10%  | 0.24 eV | 0.88 fs | 0.65 fs | 0.17 fs | 0.00268   |
| ±20%  | 0.34 eV | 1.05 fs | 0.65 fs | 0.26 fs | 0.00296   |
| ±30%  | 0.43 eV | 1.2 fs  | 0.73 fs | 0.34 fs | 0.00296   |

Settings for the kick, FWHM/RMS pulse duration, and pulse structure sample data: Kick = 22.5 eV, Pulse duration = 4.85 fs. Additional settings for the decay data: Decay = 7.0 fs.
We ran hundred experiments and averaged the results. We do not have true labels for the experimental data. In addition, we need to identify to what extent our modeled noise replicates actual SASE pulse shaping. Further steps to a successful implementation of these advanced methods involve closing the gap between simulation and experimental data through an instrument-specific treatment of the measurement noise and a reliable concept for error and reliability estimation, which we will investigate in future work.

### Output Speed
For an evaluation of the input images at the speed of the XFEL repetition rate under investigation, an efficient analysis is inevitable. NNs are known for delivering outputs quickly. We have used several batch sizes, starting from one image up to 4096 as input. Investigating this is important as such a comparison determines whether a batch-wise analysis is performing better than analyzing image-wise. Batch-wise evaluation is specifically suited for the European XFEL facility, since a train with a very fast succession of pulses is followed by a pause of several milliseconds that can be used for analysis purposes. We have tested how fast the NN output is generated on a GeForce RTX 2070 GPU (Tab. 2).

| BS   | 1   | 64  | 128 | 256 | 512 | 1024 | 2048 | 4096 |
|------|-----|-----|-----|-----|-----|------|------|------|
| Dur [ms] | 1.38 | 1.44 | 1.48 | 1.54 | 1.62 | 1.54 | 1.38 | 1.44 |

**Table 2.** Time measurements for predictions of the trained model on a GeForce RTX 2070 card with different batch sizes (BS). We ran hundred experiments and averaged the results.

The model is able to reach quick predictions mostly independent of the batch size as the computation on a GPU runs all tasks, i.e., computes a prediction for each image within the batch, in parallel. In general, the number of input images in one batch is only limited by the RAM of the used GPU. Thus, it is apparent that it is advantageous to analyze a larger batch of data than individual images. With a batch size of 4096 our current model is already able to keep up with the European XFEL in high-repetition mode for online predictions.

### Reliability Estimation
Next to fast evaluation, a degree of certainty in the NN predictions must be ensured. As shown in the results, the NN prediction may deviate quite substantially from the target. Some of the difficulties can be directly circumvented. By determining the kick, for example, we can already filter whether a prediction regarding the labels Auger decay time or pulse duration is reasonable. But this still does not give us a direct statement regarding how certain the NNs prediction is. Optimally, we would want to have a reliable measure of how good the predictions of the trained models are, even for unknown shots without a target.

There are several ways to determine the prediction uncertainties of NNs. The epistemic uncertainty determines the uncertainty due to insufficient knowledge. This can be done, for example, via Monte Carlo dropout or Monte Carlo batch normalization. The aleatoric uncertainty determines the uncertainty due to the complexity of the problem. This can be done by creating a fitted cost function. Both uncertainty determinations can be combined in one procedure as previously shown.

### Gap between Simulation and Reality
So far, our NNs are suited only for data that look exactly like the input shown in Fig. 11. Whether the NNs are suitable for predictions on experimental data (cf. Fig. 1) is not easy to validate, especially since we do not have true labels for the experimental data. In addition, we need to identify to what extent our modeled noise replicates real noise of the spectrometer, e.g., electronic ringing of the detector readout. There are two ways to tackle the gap between simulation and experiment. Either the real data must be denoised before the analysis (e.g., Denoising Autoencoders) or the simulation data must be provided with additional, appropriately modeled noise. Simultaneous approaches in both directions should give a more complete understanding for mitigating this issue in future efforts.

### Responding to Changes
We have shown that our developed NNs work on data with several values of noise. However, when utilizing real-life TOF spectrometers, it may occur that TOF sensors fail or produce unrealistic results. In such cases NN re-training or knowledge extension is inevitable. Here, online learning is a helpful tool. In this case, the model is trained continuously on newly generated data. Thus, the training can be quickly adapted to new environments. To circumvent the catastrophic forgetting of NNs, continual learning may be utilized.

### Conclusion
In this article, we demonstrated a path toward online characterization of free-electron laser pulses by applying NN on detector images captured with angular streaking. In addition to several predictable characteristics, we have been able to identify and confirm dependencies between the respective characteristics that can be used to control the machine settings during experimental campaigns. This way, the angular-streaking technique has the potential to be leveraged from the proof-of-principle stage to a robust and highly advanced diagnostic tool for all free-electron laser facilities, including high-repetition-rate operation. In addition, the live updates on X-ray pulse changes may also be used for a more detailed control of the parameters and for actual SASE pulse shaping. Further steps to a successful implementation of these advanced methods involve closing the gap between simulation and experimental data through an instrument-specific treatment of the measurement noise and a reliable concept for error and reliability estimation, which we will investigate in future work.
Methods: Machine Learning Procedure Design

In real-world XFEL experiments, the spectrograms or pulse characteristics of individual SASE pulses have to be reconstructed from the detector image. There are first approaches for deriving single-shot pulse characteristics of rapid sequences of high-repetition-rate XFELs. Unfortunately, they are only sparsely suitable for providing detailed insights via real-time online processing during experimental campaigns. Here, we apply specifically developed NNs on the angular streaking approach to demonstrate the possibility of a fast online pulse characterization, as NNs, particularly convolutional NNs, have proven to be suitable for similar challenges.

General Machine Learning Problem Formulation

For each pulse characteristic, we need to train a NN that takes detector images as inputs (cf. Fig. 10). The outputs for each of the NNs vary and are listed below:

- **Kick**  
  The kick is the amplitude of the wave within the detector image (cf. Fig. 11). When changing the kick, the spectrogram stays as is as the kick only affects the wave within the detector image. That is the reason why the kick is easily extractable from the detector images. The NN has to solve a regression task, where the output is one number in the unit eV.

- **FWHM Pulse Duration**  
  The FWHM pulse duration is well extractable from the spectrogram, as it can be seen as $2 \cdot 35 \cdot \sigma$ (cf. Eq. 3) in the direction of $x$ (time scale) with $\sigma$ being the standard deviation of the 2D Gaussian distribution in $x$ direction. The longer the FWHM pulse duration, the longer the distribution stretches in $x$ direction. Within the detector image, a change of the pulse duration affects the width of the wave. Here again, the NN has to solve a regression task, where the output is one number in the unit fs.

- **Auger Decay Time**  
  The decay affects the spectrograms as well as the detector images. Within the spectrogram, the length of the tail after the 2D Gaussian distribution indicates the decay. The longer the tail, the larger the decay. Within the detector image, a larger decay affects the distortion of the wave. The NN has to solve a regression task, where the output is one number in the unit fs.

- **Pulse Structure**  
  The pulse structure is the most challenging to extract, as the output itself consists of several values indicating the intensities of multiple spikes within the SASE pulses. By looking at the spectrogram in Fig. 11 (c), one can see that the pulse structure can be derived by summing up the intensities at each point in time along the vertical axis. The pulse characteristic will be determined here as the intensity as a function of arrival time, where the intensity is integrated over all photon energies within the 6 eV spectral bandwidth. This leads to an output similar to Fig. 4. In this case, the NN has to solve a regression task, where the output consists of several time steps in arbitrary intensity units. A note regarding the **RMS Pulse Duration**: As the RMS pulse duration can be directly derived from the pulse structure, there is no need to train an independent NN for this pulse characteristic.

After the general examination of the ML problem, the next sections will look at how the ML pipeline looks in detail and how to successively address the individual ML problems above.

Framework Conditions

In order to train NNs in a supervised manner, we require training data $\mathcal{D}_K$ of size $K \in \mathbb{N}$, which comprises $K$ simulated detector images $\mathcal{X} = \{X_i \in \mathbb{M}^{m \times n}(\mathbb{R}) \mid i = 1, \ldots, K\}$ and $K$ corresponding pulse characteristics $\mathcal{L} = \{L_i \in \mathbb{R}^L_{+} \mid i = 1, \ldots, K\}$. Here, $m$ is the number of TOF detectors used within the complete spectrometer setup and $n$ displays the electron kinetic energy in intervals. The size of $j$ changes according to the pulse characteristic that has to be predicted. In the following, we will refer to pulse characteristics as *labels*. To verify the performance of the NN, we split $\mathcal{D}_K$ into two distinct sets, $\mathcal{D}_{\text{train}}$ and $\mathcal{D}_{\text{test}}$, such that $\mathcal{D}_K = \mathcal{D}_{\text{train}} \cup \mathcal{D}_{\text{test}}$. The accuracy of the NN is determined by $\mathcal{D}_{\text{test}}$. To avoid overfitting of the NN, we utilize cross-validation.

Although we work in a simulation environment, it is reasonable to choose values that would correspond to real experimental data (cf. Fig. 1). Therefore, we take previously acquired data from earlier experimental campaigns as an example. In particular this means:
• For each of the two use cases, Ne 1s and KLL Auger electrons data, we generate a size of $K = 4.4 \cdot 10^6$ samples to be predicted. Of these, $4 \cdot 10^6$ are used for training and $4 \cdot 10^5$ for testing.

• Our angle-resolved spectrometer consists of $m = 16$ TOF detectors.

• We fix the intervals in the TOF detectors to $n = 200$, with a varying energy bin size.

More particularly, this means that the following NN architecture depends on the chosen parameters, though it can be adapted easily if, e.g., more TOF detectors are added.

**Preparing the Simulation Data**

We derive artificial detector images for Ne 1s and KLL Auger electrons from the simulation environment as introduced by Hartmann et al. The kinetic energy of the 1s photoelectrons depends on the ionizing X-ray photon energy, which, in this case, is set to 1180 eV. We include a spectral bandwidth of 6 eV for the X-ray pulses, but omit the effect of a potential chirp in these simulations, which would only have a marginal effect on the parameters reconstructed in this study. A photon energy of 1180 eV results in Ne 1s electron kinetic energies centered at $\sim 310$ eV with a standard deviation of 3 eV (according to the X-ray spectral bandwidth), while the Auger electron kinetic energies are independent of the X-ray photon energy and bandwidth, with the main peak lying at $\sim 804$ eV and a standard deviation determined by the detector resolution.

Figs. 11a and 11b, are simulated without artifacts. In 11a, there is only one randomly placed Gaussian distribution present in the spectrogram. The underlying pulse structure is neglected so far. To get closer to the real data (cf. Fig. 1), we implement three steps. We add a pulse structure and noise to the simulated detector images and prepare the data for NN training by utilizing data normalization.

**Step 1: Adding a Pulse Structure to the Spectrogram**

To achieve a SASE-like temporal structure in the spectrogram, we modulate the original Gaussian time distribution with a spiky intensity profile (cf. Fig. 4). We obtain the latter by generating a comb of Gaussian spikes with randomized amplitudes and spike durations as predicted by theory for a typical setting of an XFEL in ultrashort-pulse mode.

**Step 2: Adding Noise to the Detector Image**

Additional noise is added to $X \in \mathcal{I}$ during training and testing as shown in Eq. 4. A given percentage $p$ of the maximum intensity value $x_{\text{max}}$ of $X \in \mathcal{I}$ is used as an upper and lower bound of an equal distribution $\mathcal{G}$ to draw $w$ from:

$$X_{\text{noisy}} = (x_{i,j} + (x_{\text{max}} \cdot w))_{i=1,\ldots,m,j=1,\ldots,n}, w \sim \mathcal{G}(-p,p), \quad (4)$$

Figure 12 displays a detector image with different levels ($p \sim [0.0, 0.3]$) of added noise.

**Step 3: Normalizing the Data**

It is evident, that the range of the intensity values differs from case to case. To counteract this, we perform a min-max-normalization for each $X \in \mathcal{I}$. Therefore, the minimum ($x_{\text{min}}$) and maximum ($x_{\text{max}}$) intensity value of $X$ are used to perform the transformation for each pixel value $x_{k,l}$:

$$X_{\text{norm}} = \left( \frac{x_{k,l} - x_{\text{min}}}{x_{\text{max}} - x_{\text{min}}} \right)_{k=1,\ldots,m,j=1,\ldots,n}, \quad (5)$$

After normalization, all values in $X_{\text{norm}}$ lie within the interval $[0,1]$. The data displayed in Fig. 11 have not yet been normalized.

**Designing the Machine Learning Models**

As we want to extract information from images, the most intuitive solution is to use a convolutional NN, which uses convolutions and pooling to extract low- and high-level features such as edges and predicts an estimate of those features using fully-connected layers. In our case, the estimates ideally should correspond to the target pulse characteristics.

**Architecture**

One key problem is the dimensions of the detector images, which are not equal in size and therefore not symmetrical. This fact was taken into account in the design of the NN. The most suitable network architecture for our problem is an NN with three convolutional blocks (cf. Fig. 13). Each block contains a convolutional layer, followed by an activation function and a max-pooling layer. The convolutional layers use a 3x3 kernel, stride of 1x1 and 1x1 zero-padding. The pooling layers use a 3x3 kernel, stride of 2x2 and 1x1 zero-padding. The NN is specifically designed to cut both dimensions, i.e., width and height of the
Figure 11. Simulated Ne 1s data before [(a) and (b)] and after [(c) and (d)] adding a pulse structure. (e) and (f) display simulated angularly streaked Auger electron spectrogram and detector images, respectively, after adding a pulse structure.
Detector image without noise. (a) Detector image after adding \( \pm 30\% \) noise. (b)

**Figure 12.** Simulated detector image before (a) and after (b) adding noise.

image, in half after each block. A filter size of \([16, 32, 64]\) for the respective convolutional layers has proven to be sufficient. For the fully-connected stage (except the last layer), we use three layers with \([3200, 1600, 800]\) neurons, respectively. The size of the last layer depends on the label to predict, i.e., the size of \( j \) in \( \mathcal{L} \).

![Conv2d + ReLU + Pooling](image1)

![Fully-Connected Layer + ReLU](image2)

![Fully-Connected Layer](image3)

**Figure 13.** The convolutional neural network architecture used in the present approach. The dimensions of the first layers (orange) are displayed in [channel, height, width]. The dimensions of the fully-connected layers (violet) show the number of used neurons.

When predicting the *kick*, *FWHM pulse duration*, or *decay time*, \( j = 1 \). When predicting the *pulse structure*, \( j \) corresponds to the dimension of the spectrograms’ x-axis. We utilize the mean squared error loss function to train and optimize the network as the prediction of the pulse characteristics is a regression task in all cases.

**Hyperparameter Optimization**
The NN architecture is not the only choice to be considered. Especially when training the NNs, appropriately chosen hyperparameters are important to achieve efficient and goal-oriented training. Important parameters in this context are the *batch size*, *type of activation function*, *optimizer*, and *learning rate*. To find the best suitable combination of hyperparameters, we performed a grid-search on distinct data sets using the approach from before with the following values:

- **Batch size**: \([64, 128, 256, 512, 1024]\)
• Activation function: [ReLU, Sigmoid]
• Optimizer: [Adam, SGD (with Momentum)]
• Learning rate: [0.01, 0.001, 0.0001, 0.00001]

After NN training, we evaluated the respective parameter combinations according to the following criteria:

• Criterion 1: The test loss (after inputting $D_{\text{test}}$ into the trained NN) should be minimal.

• Criterion 2: The standard deviation of the test loss curve should be minimal to penalize slow convergence and overfitting.

In general, it should be noted that there is not only one combination of hyperparameters that achieves good results during training. Nevertheless, there has been an evident leader. The best hyperparameter configuration for all labels is a batch size of 64, a ReLU activation function, a learning rate of 0.0001, and Adam as optimizer.

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Additional information

Data and Code Availability A repository containing the detector image analysis software and according simulation data will be provided on request;
Competing interests The authors declare no competing interests.