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Femtosecond X-ray Pulse Length Characterization at the LCLS Free Electron Laser

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Abstract.

Two-color, single-shot time-of-flight electron spectroscopy of atomic neon was employed at the Linear Coherent Light Source (LCLS) to measure the laser-assisted Auger decay in the X-ray regime. This X-ray-optical cross correlation technique provides a straightforward, non-invasive and on-line means of determining the duration of femtosecond (> 40 fs) X-ray pulses. In combination with a theoretical model of the process based on the soft-photon approximation, we were able to obtain the LCLS pulse duration and to extract a mean value of the temporal jitter between the optical pulses from a synchronized Ti-Sapphire laser and X-ray pulses from the LCLS. We find that the experimentally determined values are systematically smaller than the length of the electron bunches. Nominal electron pulse durations of 175 and 75 fs yield X-ray pulse shapes of 120 ± 20 fs FWHM and an upper limit of 40 ± 20 fs FWHM, respectively. Simulations of the SASE process using the GENESIS code agree well with the experimental results.

1. Introduction

The Linear Coherent Light Source (LCLS) started lasing in the X-ray regime (0.8 – 8 keV) in the summer of 2009 with an unprecedented flux in the range of 10^{13} photons per pulse and controllable duration from a few-fs up to $\simeq 300$ fs [1]. Combined with appropriate focusing optics, peak irradiance levels of more than 10^{17} W/cm² are possible, extending multi-photon [2, 3, 4] and non-linear [5] spectroscopy into the X-ray region of the electromagnetic spectrum. In addition, with temporally synchronized ultra-fast lasers, new pump-probe experiments involving intense laser fields of radically different photon energies can be performed [6, 7]. In all experiments, utilizing both single color pulses and pump-and-probe methodologies, the X-ray pulse duration is a pivotal parameter, and so the continuous development of reliable techniques to determine it are crucial for the full exploitation of ultra-fast X-ray free electron lasers (FEL). Hence we set out to measure it in a two-color gas phase cross-correlation experiment, which meets the dual desirable criteria of being non-invasive and potentially employable as an on-line diagnostic. Our focus here is on the measurement of X-ray pulse durations in excess of 40 fs. Experiments to determine even shorter pulse durations require a completely different methodology and will be published separately [8]. In addition to the pulse length, the technique reported here can also reveal and quantify the timing jitter of the FEL with respect to an external synchronized optical laser, a parameter especially important for time-resolved pump-and-probe experiments.

A two-color process, sensitive to the separation between X-ray and near infrared (NIR) laser pulses is shown in Fig. 1. When electrons, generated by the X-ray photoionization of atoms emerge into an intense NIR field they exchange photons with the field causing *sidebands* to appear in the electron kinetic energy spectrum, yielding new lines separated from the main photoelectron line by multiples of $\hbar\omega_{NIR}$ [9, 10]. However, in the soft X-ray energy regime (1 keV) the photon energy of the NIR laser (1.55 eV) is substantially less than the natural bandwidth of the FEL (0.5 – 1 % FWHM). Therefore, it is advantageous to use the free electrons created in an Auger decay process following inner-shell excitation (resonant Auger decay) or direct ionization (normal Auger decay). Coined *laser-assisted* Auger decay (LAAD) this effect was first observed for the argon $L_{23}M_{23}M_{23}$ Auger electrons centered at 240 eV [11]. In neon the inner-shell photoionization step $\text{Ne } 1s^2 + \hbar\omega_{FEL} \rightarrow \text{Ne } 1s^1 + \epsilon l$ is followed by the filling of the Ne K-shell core-hole vacancy, which results in the Auger decay of an electron with a kinetic energy 804.3 eV [12]. Due to the narrow, fixed spectral bandwidth of the Auger line (0.23 eV FWHM), corresponding to the very short lifetime of 2.4 fs, the sidebands in the two-color Auger spectrum will be, in principle, better separated than in case of the dressed Ne $1s^{-1}$ photoelectron line. Hence, we use LAAD as a cross-correlation signature to determine the X-ray pulse duration for two different settings of the accelerator, corresponding to nominal electron bunch durations of 75 fs and 175 fs, respectively.

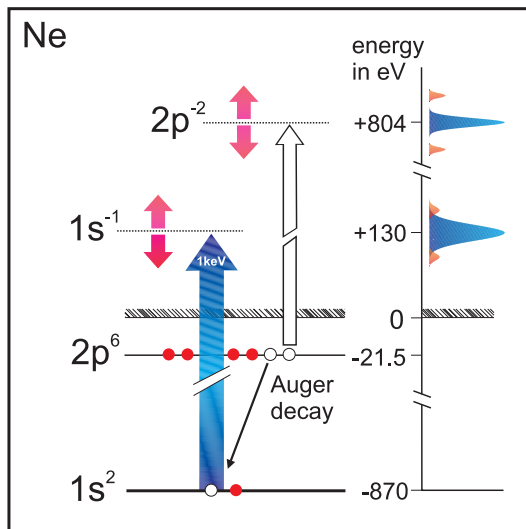


Figure 1. Simplified energy level diagram and two-color excitation schemes for atomic Ne. Note that for the Auger process the ejection of the 2p electron in the Ne^+ ion requires 44.2 eV.

2. Experimental Setup

In this work we used the diagnostic chamber of the atomic, molecular, and optical (AMO) end station. It comprises a magnetic bottle electron spectrometer (MBES), a gas inlet providing an effusive gas flow at the interaction region, and various beam imaging tools [13]. After the interaction of neon gas with the X-ray beam, the resulting photo- and Auger-electrons are analyzed by the MBES with an angular acceptance of approximately 0.8 sr (30° half-angle) at electron kinetic energies around 0.8 keV [14]. The performance of the MBES is briefly demonstrated in Fig. 2, which shows the main features of the electron spectrum recorded after exposure of Ne atoms to X-ray pulses with a mean photon energy of 990 eV. In the lower kinetic energy region, the main Ne $1s^{-1}$ photo-line appears at 120 eV. The spectrum shown in Fig. 2 (a) is an average over 200 single-shot spectra and the line profile is well represented by a Gaussian fit of 7 eV FWHM. This value is mainly determined by the natural bandwidth of the LCLS. To demonstrate the photon energy fluctuations from shot to shot, two single-shot spectra are shown in Fig. 2 (a) demonstrating the single-shot capability of the MBES. This is a necessary prerequisite for experiments with the strongly varying temporal, spatial and spectral beam conditions inherent in FEL radiation.

In the energy region around 770 – 810 eV the Ne KLL Auger spectrum is presented (Fig. 2 (b)). For the MBES only the most intense line around 804 eV kinetic energy is shown, corresponding to the transition to the $\text{Ne}^{2+} 3p^4 \ ^1D$ final state. For comparison, we also display a spectrum recorded with the high-resolution electron time-of-flight (ETOF) spectrometer situated in the AMO high-field physics (HFP) chamber demonstrating that no additional structures show up on the high energy side of the 1D line. Since the width of the Auger lines does not depend on the energy width of the

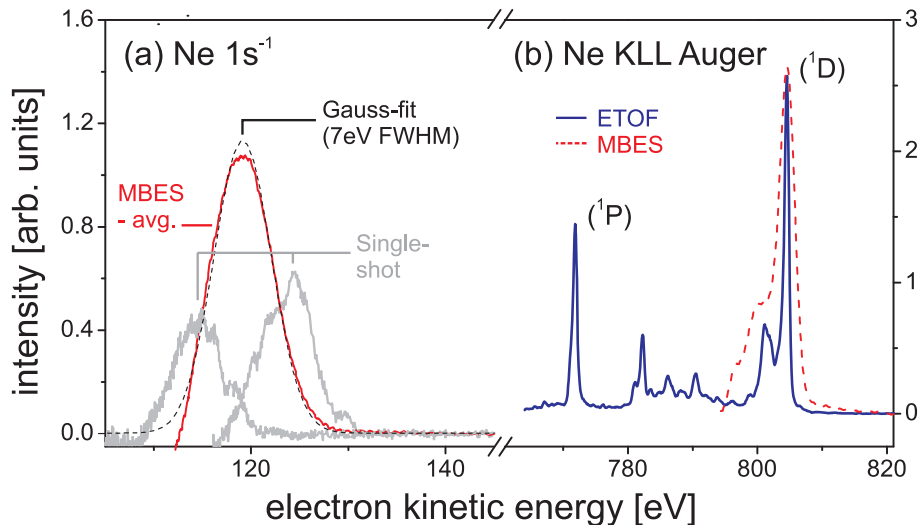


Figure 2. Photo- and Auger-electron spectra from neon after irradiation by 1 keV LCLS pulses. (a) Averaged $\text{Ne } 1s^{-1}$ photo-line situated at ~ 120 eV: the experimental line is described by a Gaussian profile of 7 eV FWHM corresponding to the natural FEL bandwidth. Single-shot spectra (not to scale) highlight the shot-to-shot energy jitter of the FEL. (b) Ne KLL Auger spectrum obtained with the MBES averaged over 200 shots: the line width of the main ${}^1\text{D}$ line is 3 eV FWHM (dashed-line). The full Auger spectrum recorded with an ETOF spectrometer in the HFP-chamber is shown for comparison (solid-line).

exciting photon pulse or on its spectral jitter, the spectra allow us to determine the energy resolution of the analyzers. An electron energy resolution of 0.4% (~ 3.0 eV FWHM) was achieved with the MBES by carefully optimizing the retardation voltages and acceptance geometry of the spectrometer [14]. The high-resolution spectrum obtained by using the ETOF is measured with an energy resolution $\Delta E/E$ of the order of 0.1%. However, the narrow angular acceptance of the ETOF (0.007 sr) does not permit single-shot acquisition with sufficient statistics.

For the measurements presented here the LCLS was operated in the 250 pC (charge per electron bunch) mode, at a pulse repetition frequency of 30 Hz and an electron beam energy of 4.7 GeV ($\hbar\omega_{FEL} = 1.0$ keV nominal). Two different compression settings of the accelerator were used in order to achieve electron pulse lengths of $\tau_{el} = 75$ fs and 175 fs resulting in peak currents of 1.79 kA and 0.91 kA, respectively [1]. The total X-ray energy per pulse typically varied from 0.5 – 2 mJ with fluctuations of 20% (FWHM) about the mean energy observed from the pulse energy distribution, which was monitored by a photon flux detection setup integrated into the beam line [15]. Adaptive focusing mirrors, arranged in a Kirkpatrick-Baez geometry, were adjusted to produce a FEL spot diameter of about $60 \mu\text{m}$ in the interaction region. The NIR laser system delivered a pulse of duration $\tau_{NIR} = 100$ fs (FWHM) at a central wavelength of 800 nm and a pulse energy of $200 \mu\text{J}$ at a pulse repetition frequency of up to 360 Hz. The last turning mirror for the laser, located on the FEL beam axis inside the vacuum chamber, has a 2 mm diameter hole in its center, through which the focused FEL beam was directed. A NIR

spot of $300\ \mu\text{m}$ FWHM ensured good spatial overlap of the NIR and X-ray beams and yielded focal intensities of the NIR laser $I_{NIR} \sim 10^{12}\ \text{Wcm}^{-2}$ in the interaction region. Hence, the experiment was quite insensitive to any shot to shot jitter in the spatial overlap due to beam pointing fluctuations in either the Ti-sapphire or free electron laser. The spatial overlap between the LCLS and NIR laser beams was controlled by introducing a Ce:YAG viewing screen into the region of interaction and adjusting the NIR laser beam steering such that both spots were overlapped. Coarse timing (with a precision of ~ 30 ps) was achieved with a fast photodiode. Final optimization of the temporal and the spatial overlap was done by monitoring the LAAD yield itself. Details on the laser system and its synchronization to the LCLS accelerator can be found in Ref. [6].

3. Results and Discussion

A set of electron spectra recorded as function of the relative delay time (in the range of ± 1 ps) between the NIR and X-ray pulses is shown in Fig. 3. For clarity, only the main Auger line and the part of the spectrum lying to its high energy side are displayed. Each spectrum represents the acquisition of many single-shot spectra ($\sim 10^3$), which were sorted a posteriori into 50 fs time bins, according to the LCLS phase cavity electron arrival time monitor, and averaged [6]. In the cross-correlation region the action of the NIR field causes a strong redistribution of the Auger electrons, resulting in the evolution of a prominent feature at higher kinetic energies and a concomitant intensity reduction of the main line at 804.3 eV. The total electron yield remains constant, as the NIR field does not contribute to the primary ionization process. Since the current setup cannot resolve individual sidebands, the LAAD process appears as a rather coarse broadening of the Auger line. Nevertheless, we can infer from the cut-off in the kinetic energy spectrum at maximum overlap ($\tau_{\delta t} = 0$) that up to *fifteen* NIR photons are absorbed from the dressing field. This is in good agreement with the so-called Simpleman's model [11]. Here, the final kinetic energy U of an electron introduced into a moderate NIR field (with wavelength λ_{NIR}) with an intensity I_{NIR} is given by:

$$U = U_a - \sin(\phi_b) \cos(\theta_b) \sqrt{8U_a U_p} \quad (1)$$

ϕ_b is the phase of the NIR-field at the instant of ionization, θ_b is the angle between the light polarization and the initial velocity of the electron at birth and U_a is the initial kinetic energy. U_p is the instantaneous ponderomotive energy at the moment of birth:

$$U_p[\text{eV}] = 9.3310^{-14} \times \lambda_{NIR}^2[\mu\text{m}^2] \times I_{NIR}[\text{W}/\text{cm}^2] \quad (2)$$

For $\lambda_{NIR} = 0.8\ \mu\text{m}$ and $I_{NIR} = 1.2 \times 10^{12}\ \text{Wcm}^{-2}$, U_p results in 0.07 eV. Which, when substituted into Eq. 1 gives a maximum energy $U \simeq 826$ eV or, in other words, the absorption of about 14 NIR photons from the field. This is in excellent agreement with the experimental findings shown in Fig. 3. For a given intensity and wavelength of the NIR laser, U scales with the square root of the initial kinetic energy U_a . Hence,

compared to two-color processes previously observed at extreme ultra-violet wavelengths the high number of sidebands observed here at low NIR intensity is a direct consequence of the high kinetic energy of the Auger electron [10, 16].

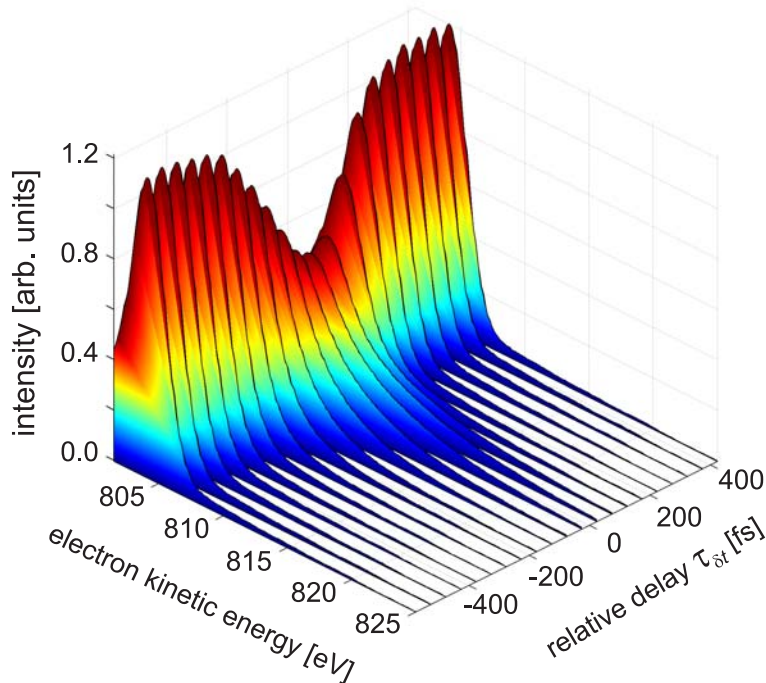


Figure 3. Neon Auger electron energy spectra as function of the relative delay time $\tau_{\delta t}$ between the NIR ($\tau_{NIR} = 100$ fs) and the X-ray pulses ($\tau_{el} = 75$ fs). Around the relative zero-point the strong depletion and broadening of the main line corresponds to LAAD.

In Fig. 4 several single-shot electron spectra, recorded at the nominal maximum temporal overlap between the X-ray and NIR pulses, are shown. The strong shot-to-shot variation in the LAAD component demonstrates clearly the timing jitter in the synchronization and highlights the usefulness of the LAAD effect as a sensitive monitor of the temporal overlap of the pulses. This is even more evident when the experimental line profiles are modeled with a simplified analytical model based on the so-called soft-photon approximation (SPA) allowing us to simulate the LAAD spectrum [17]. In general, the SPA gives excellent agreement with LAAD data and can match the more elaborate non-perturbative methods such as the Time Dependent Schroedinger Equation (TDSE) when the electron kinetic energy is large compared to the dressing photon energy [17, 18].

The simulated LAAD spectra are defined by four parameters: the NIR laser pulse duration, τ_{NIR} and intensity I_{NIR} , the FEL pulse duration (and shape) τ_{FEL} and, finally, the relative temporal delay between the two pulses $\tau_{\delta t}$. The NIR pulse width was measured with an auto-correlator to be $\tau_{NIR} = 100$ fs (FWHM with Gaussian profile). Monitoring the pulse energy and the focal spot size, a peak intensity of $I_{NIR} = 1.2 \times 10^{12}$ Wcm $^{-2}$ was estimated. The average jitter on the relative temporal delay was

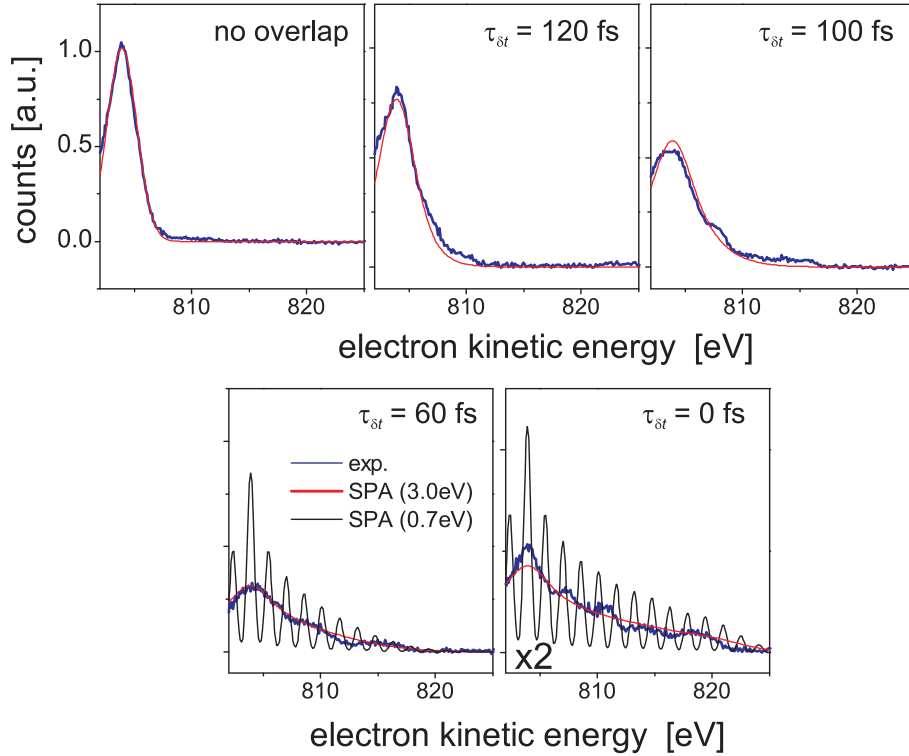


Figure 4. Selected single-shot (blue line) LAAD spectra ($\tau_{el} = 75$ fs) and corresponding results from the soft-photon approximation. The SPA spectra were calculated for $\tau_{NIR} = 100$ fs FWHM ($I_{NIR} = 1200$ GW/cm²) and for $\tau_{FEL} = 40$ fs. Shown are spectra simulated with the experimental energy resolution of 3.0 eV FWHM (red line) as well as with 0.7 eV FWHM (gray line in the lower two figures) in order to demonstrate the underlying sideband structure, which is otherwise washed out by the energy resolution of the MBES.

determined with two independent approaches: first, we analyzed the cross-correlation width for FEL pulse duration settings of ~ 20 fs or shorter pulses (so-called 20 pC mode). Here the X-ray pulse duration is negligible in the determination of the correlation width which is only determined by the optical pulse duration and the jitter. Knowing the laser pulse duration, we obtain from the LAAD analysis a value $\tau_{jitter} = (140 \pm 20)$ fs FWHM including the electron pulse arrival time correction data from the phase cavity measurement [6]. An alternative approach is to determine the jitter from the single-shot spectra by fitting the data as shown in Fig. 4 with the SPA simulation. Here the delay between the NIR and the X-ray pulse was used as a fit parameter yielding a list of time delays and thus the average jitter. The calculation was performed with X-ray pulse durations $\tau_{FEL} = 75$ fs (FWHM), as the upper limit for the expected X-ray pulse duration, and also for $\tau_{FEL} = 40$ fs (FWHM), as determined in this work (see below). The jitter determined for both pulse durations agrees within the error bars with the value determined via the cross-correlation approach, i.e. $\tau_{jitter} = (140 \pm 20)$ fs FWHM. We note that the jitter depends on many parameters and is sensitive to minor changes in the settings of the feedback electronics, the beam transport optics and associated systems,

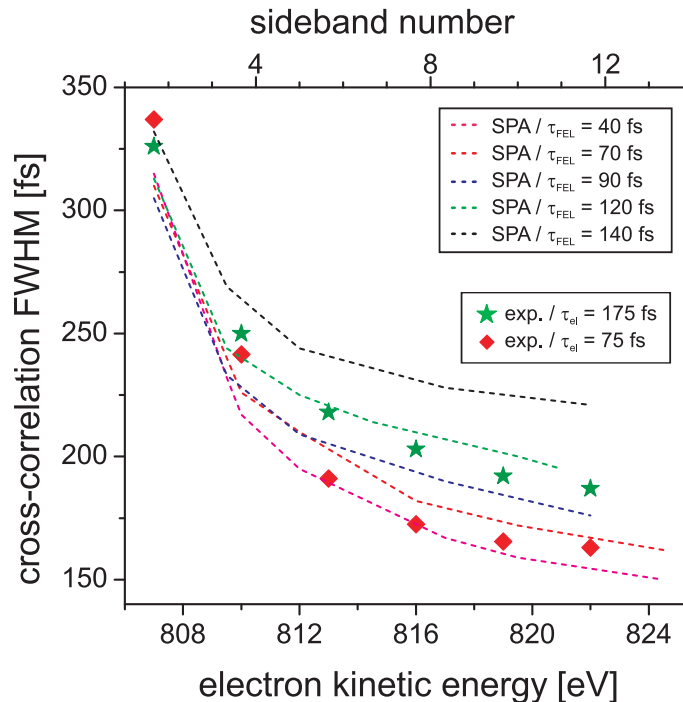


Figure 5. Comparison of the experimentally obtained cross-correlation widths with those calculated within the soft-photon approximation using the following parameters (in FWHM): $\tau_{FEL} = 40 - 140$ fs, $\tau_{NIR} = 100$ fs, $\tau_{jitter} = 140$ fs, $I_{NIR} = 1.2 \times 10^{12}$ Wcm $^{-2}$.

distribution system and mechanical vibrations. For example, other investigators have reported values approaching 300 fs (FWHM) [6].

As discussed above, under our experimental conditions, LAAD is a highly non-linear multi-photon effect, which has a strong dependency on the driving NIR laser intensity. Thus, the cross-correlation width at these intensities is no longer a direct measure of the cross-correlation of the laser and the X-ray pulse. Therefore, quantitative information about the X-ray pulse duration τ_{FEL} can only be extracted by comparing the LAAD features to the theoretical model. In order to simulate the experimental data using the soft-photon model the NIR laser pulse, modeled as a Gaussian temporal profile, is divided into 5 fs time windows with constant intensity. Within each such segment the resulting LAAD spectrum, including all amplitudes from up to 30 sidebands, is calculated successively. Since the NIR laser does not contribute to the initial photoionization step, the relative amplitude of each segment-spectrum is given by the FEL photon flux only. Here, the approximate pulse shapes from the FEL simulation were used as input (see below). In the final step the component spectra were summed to represent an experimental single-shot spectrum. To simulate the full experimental set as shown in Fig. 3, a delay scan from -500 fs to +500 fs in 10 fs steps is used. For each time step, 50 single-shot spectra were accumulated and a random additional temporal time difference was introduced to simulate the time jitter. According to the experimental findings, a Gaussian time delay distribution with a FWHM of 140 fs was used.

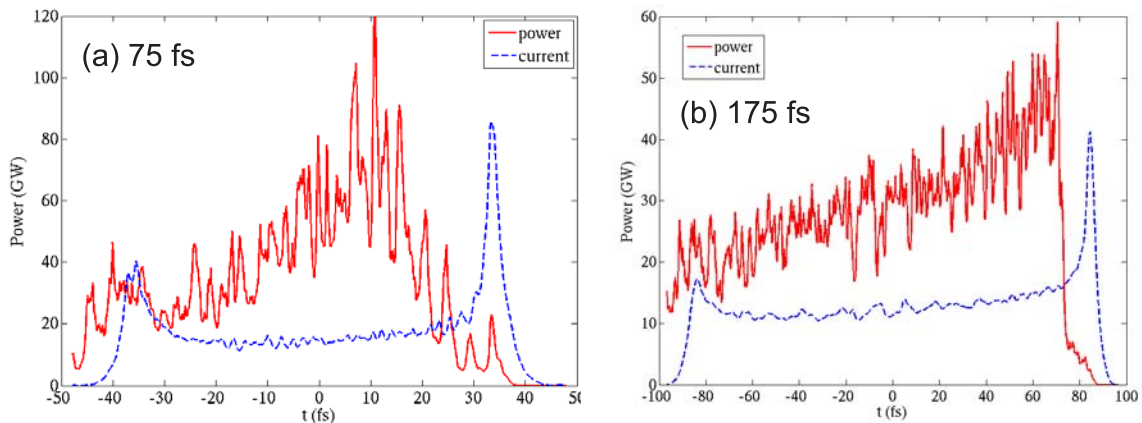


Figure 6. Shown is the temporal profile of GENESIS-simulated FEL pulses. The FEL radiation power in GW (red line) and the electron current profile (blue curve) are shown for (a) $\tau_{el} = 75$ fs and (b) for $\tau_{el} = 175$ fs. The bunch head is at the left.

Shown in Fig. 5 is a comparison of the experimental cross-correlation widths, obtained by a projection along the time axis (cf. Fig. 3), and the results of the SPA simulation. As expected, the calculated widths are strongly dependent on the NIR intensity, i.e. for low order sidebands they are considerably greater than for higher sidebands. The experimental widths extracted from the cross-correlation curves recorded with $\tau_{el} = 75$ fs and 175 fs respectively show the same general behavior as a function of kinetic energy and can well be described by the SPA simulation. A closer inspection of Fig. 5 shows that the measured pulse durations for $\tau_{el} = 75$ fs point to an actual width of $\tau_{FEL} = (40 \pm 20)$ fs, which is also in agreement with indirect measurements presented in ref. [4]. The experimental values obtained for $\tau_{el} = 175$ fs are best described by a pulse of width $\tau_{FEL} = 120 \pm 20$ fs (FWHM). These results of the LAAD cross-correlation analysis are further corroborated by a set of FEL simulations using the GENESIS code [19]. The radiation power profiles shown in Fig 6 are averaged over 50 Genesis runs using random initial shot noise. For the experimental parameters chosen GENESIS predicts for an electron pulse duration of $\tau_{el} = 75$ fs an almost Gaussian-like X-ray pulse with $\tau_{FEL} \simeq 40$ fs (FWHM) and for $\tau_{el} = 175$ fs a more triangular-like X-ray pulse with a total width of 170 fs (120 fs FWHM). It is also noteworthy that a 120 fs FWHM Gaussian X-ray pulse yields results similar to a triangular pulse. This is due to the fact that the jitter and optical pulse duration dominate the cross-correlation and thus the X-ray pulse *shape* cannot be determined directly. Besides the good agreement between experimentally determined and simulated values for nominal pulse durations of 75 and 175 fs, first measurements for the longest (nominal 300 fs) pulses yield a much shorter pulse duration of only ~ 120 fs FWHM, whereas the GENESIS simulation suggest a 300 fs long X-ray pulse. Further studies are needed here to resolve this discrepancy for the longest FEL pulses.

4. Conclusion

In conclusion, we have analyzed the pulse width and timing performance of the LCLS X-ray free electron laser using single-shot time-of-flight electron spectroscopy to perform an X-ray-optical cross-correlation measurement at the LCLS. We have investigated for the first time the two-color laser assisted Auger decay process in the X-ray region, demonstrating a good agreement between the experimental and simulated LAAD spectra. Also, the results show that two-color processes, known from extreme ultraviolet FEL sources, can be directly transferred to the X-ray regime and are even more pronounced at the resulting electron energies. The corresponding cross-correlation analysis shows for the tested electron pulse durations of $\tau_{el} = 75$ fs and 175 fs that the effective X-ray pulses (in FWHM) are shorter than the electron bunch and values of $\tau_{FEL} = 40$ fs and 120 fs (FWHM) respectively, have been found. Furthermore, our analysis of the single-shot statistics demonstrates that synchronization between the NIR and X-ray lasers can be achieved with a precision of 140 fs (FWHM). Finally, we note that with shorter optical pulses and a lower jitter, this cross-correlation method has the potential to provide accurate on-line information not only on the width of shorter X-ray pulses, but also on the (average) pulse-*form* of the LCLS radiation itself.

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