Spectroscopic Characterization of VUV Free Electron Laser Pulses


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Due to the stochastic nature of Self Amplified Spontaneous Emission (SASE) it is crucial to measure for single pulses the spectral characteristics of ultrashort pulses from the Vacuum-UV Free Electron Laser (VUV-FEL) at DESY. To meet this particular challenge, we have employed both photon and photoelectron spectroscopy. Each FEL pulse is composed of an intense and spectrally complex fundamental, centered at a photon energy of about 38.5 eV, with a bandwidth of 0.5% accompanied by higher harmonics each carrying an intensity of typically 0.3 to 0.6% of that of the fundamental. The correlation between the harmonics and the fundamental is in remarkable agreement with a simple statistical model of SASE FEL radiation.© 2006 Optical Society of America

Free Electron Lasers, operating on the principle of Self Amplified Spontaneous Emission, open up completely new vistas in intense laser matter interaction as they have the potential to provide ultrashort pulses of coherent laser radiation with photon energies far above the ionization thresholds of matter. As a result, non-linear optics and spectroscopy can be extended into the vacuum ultraviolet (VUV) region of the electromagnetic spectrum for the first time. In contrast to traditional visible lasers, the predominant interaction of VUV-FEL radiation is expected to be with inner shell electrons. Hence new physical phenomena, in which electrons in resonant autoionizing and continuum states play an important role, are expected to be observed.

Established VUV sources, such as storage ring based insertion devices and high harmonics of optical lasers, lack the enormous brilliance of the FEL, but they do enjoy stable conditions of operation. In contrast, the statistical character of the SASE process gives rise to intensity and frequency fluctuations for each individual shot. Hence, it is absolutely essential to have the capacity to determine the salient properties of the FEL beam, such as spectral distribution and intensity variations, on a shot-to-shot basis. Similar to SASE FELs in the visible frequency range, SASE VUV-FELs emit quite intense odd and even harmonics up to the percent level. The ratio of the 3rd harmonic to fundamental intensity grows strongly within the linear regime of operation and is limited to a theoretical maximum of 2% when the FEL output reaches saturation. To comprehensively characterize the FEL spectral distribution, we combined a high resolution grazing incidence spectrometer to record single shot spectra, with a time of flight photoelectron spectrometer capable of recording electrons produced by fundamental, as well as harmonics, of the FEL radiation within one single pulse.

The kinetic energy analysis of the photoelectrons was obtained by a magnetic bottle electron analyzer similar to that described in ref. Electrons produced by the interaction of an effusive gas jet and the FEL beam (focus ∼ 30 μm FWHM) in the acceptance volume of the analyzer are directed by a strong permanent magnet (0.5 T) towards a 65 cm long time-of-flight (TOF) tube. A solenoid (50 mT) then guides the electrons further. The total collection efficiency of this configuration is close to 100% for all considered kinetic energies. The total photoelectron current was detected with a conventional two-stage channel plate detector. A typical energy resolution of 5% of the kinetic energy was achieved, which can be improved by decelerating the
electrons with the aid of an electrostatic retardation field at the entrance of the TOF. Rare gases (He, Ar and Xe) were introduced through a capillary of 500 µm inner diameter. The background pressure was approximately $2 \times 10^{-8}$ mbar without gas, and was raised to $2 \times 10^{-7}$ mbar with a rare gas. The linear accelerator was operated with a repetition frequency of 2 Hz for the 'macro' pulse, consisting of a pulse train of up to 20 individual electron bunches, with 1 µs separation. The central wavelength of the SASE FEL radiation was about 32.2 nm (38.5 eV) with a temporal width, estimated from the spectral properties of each pulse to be less than 50 fs. The average energy of the pulses, monitored by detectors provided by DESY varied typically between 5 and 20 µJ per pulse. These values correspond to $\sim 10^{12}$–$10^{13}$ photons per pulse.

A typical photoelectron spectrum recorded by summing over 256 individual FEL pulses is displayed for xenon in figure 1. The strongest feature in the spectrum corresponds to the photoionization of the two spin-orbit components of the Xe 5p shell ($2^2\Pi_{3/2}$ and $2\Pi_{1/2}$ at kinetic energies of $E_{\text{kin}} = 26.4$ and 25.1 eV, respectively). Additional structures at kinetic energies $E_{\text{kin}} > 30$ eV are attributed to the photoionization of the Xe atoms in the 5p, 5s and 4d shell by the second ($\hbar \omega$ (2nd) = 77 eV) and third harmonic ($\hbar \omega$ (3rd) = 115.5 eV) of the fundamental FEL radiation. In particular, due to the high 4d photoionization cross section the broad peak at $E_{\text{kin}} = 30$ eV is clearly visible. It should be stressed that we cannot distinguish between photoionization induced by the higher harmonics of the FEL and direct two- and three-photon ionization, which should become possible for high intensities. We have excluded such non-linear processes, since almost identical values are deduced from the spectra of all three gases. Since the energy dependence of the partial photoionization cross section for the Xe 5p, 5s and 4d shell is known with high accuracy, the integrated intensity of the corresponding lines in the photoelectron spectrum can directly be converted to intensities of the harmonics relative to the fundamental. Average values, for both the 2nd and 3rd harmonics, were found to be equivalent within the error bars. Ratios of (0.3 ± 0.2)% and (0.6 ± 0.2)% have been determined for ~5 µJ and ~20 µJ FEL pulse energies, respectively.

Due to the inherent statistical fluctuations in the SASE process, there may be considerably more harmonic content in an individual FEL pulse than the mean values given above. This can be addressed by measuring photoelectron spectra for each single FEL pulse. Due to the large photon flux per pulse and high rare gas photoionization cross sections single shot spectra were easily recorded. In figure 2 the absolute intensity of the Xe 4d$^{-1}$ ($E_{\text{kin}} = 47$ eV) line vs. the 5p$^{-1}$ ($E_{\text{kin}} = 25$–26 eV) line is shown, i.e. indicating the relative intensity of the 3rd harmonic of the FEL with respect to the intensity of the fundamental. The plot shows a strong correlation between a high FEL intensity and a strong 3rd harmonic signal. For intense pulses the spread of the data points is quite large. This can be explained by the strong dependence of the 3rd harmonic on intensity spiking in the fundamental, since the intensity of the 3rd harmonic is proportional to the cube of the intensity of the fundamental $I_3(t) \propto [I_1(t)]^3$.

To get a better understanding a simple theoretical approach was used to simulate correlations between fundamental and harmonic pulse energies (see figure 3). In the underlying model we assumed that the SASE FEL radiation can be described in the “linear” mode of operation, i.e. exponential gain regime, using Gaussian statistics. This analytical approach is not valid.

![Figure 1](image1.png)  
**Fig. 1.** Photoelectron spectrum of Xenon recorded after interaction with VUV photons from the FEL lasing at 32.2 nm (38.5 eV). The 5p photolines as well as the additional structures induced by the 2nd and 3rd harmonics of the FEL are indicated.

![Figure 2](image2.png)  
**Fig. 2.** The relative pulse energies for the fundamental and the 3rd harmonic were measured for 100 individual FEL pulses (stars) by TOF spectroscopy. The simulated SASE pulses (6 modes, circles) and the TOF data were scaled to the provided average pulse energy of ~5 µJ.
in the “non-linear” regime, at saturation. In our case where the FEL operates between both regimes, the qualitative, linear approach still represents the fundamental properties well. In the linear region, the instantaneous intensity of the fundamental $I_1(t)$ shows a negative exponential probability distribution $p(I_1(t)) = \langle I_1(t) \rangle \cdot \exp(-I_1(t) / \langle I_1(t) \rangle)$, where $\langle \rangle$ denotes the average over an ensemble of pulses. The summation over M sub-pulses yields a reference for one SASE pulse at the fundamental wavelength. $M$ can simply be measured by analyzing the energy fluctuation data acquired from the fundamental radiation. However, summing the cube of the instantaneous intensity for each sub pulse represents a relative value for the 3rd harmonic pulse. To compare the simulated data to the experiment, the average ratio of the simulated 3rd harmonic to the fundamental was scaled to the experimentally determined average. With such a model, we can analyze the individual temporal pulse shapes for all simulated pulses in figure 2. Indeed, a high ratio of the 3rd harmonic to the fundamental for one single FEL pulse corresponds to a distinct spike in the time domain of the fundamental and, even more pronounced, in the 3rd harmonic pulse. Since the model yields good qualitative agreement with the experiment, we infer that the underlying temporal structure predicted by it must also be a good qualitative representation of the real pulse shapes which we cannot measure directly.

To investigate the spectral distribution in more detail, high resolution single-shot spectra (figure 3) were recorded in the range from 10–40 nm by a grazing incidence stigmatic spectrometer ($\Delta \lambda \sim 0.04$ nm @ 30 nm). The spectrum is acquired on a fluorescent screen (spectral window of $\sim 2$ nm), which is imaged by a gated intensified camera capable of recording spectra from single FEL pulses out of a bunch train. Spectra of the 2nd and the 3rd harmonic, averaged over 2500 shots, have been recorded (see inset in figure 3). Considering all spectrometer components the wavelength dependent response was calculated. The relative intensities of the harmonics to the fundamental are determined as $2^{nd}/1^{st} = (0.35 \pm 0.35 \%)$ and $3^{rd}/1^{st} = (0.4 \pm 0.4 \%)$ by the spectrometer for an average pulse energy of $\sim 20 \mu J$. Despite the different location of the detectors, these values are in good agreement with the photoelectron measurement.

In conclusion, we have characterized the spectral properties of the VUV-FEL lasing at a wavelength of 32.2 nm. Using time of flight photoelectron spectroscopy, the ratio of the pulse energy of the fundamental and the two higher harmonics within one FEL pulse have been measured on a shot-to-shot basis (1 µs pulse spacing). Our detection method is non-perturbative, thus it can be used as an online detector for experiments to deliver information about the harmonic content of the FEL pulses.

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14. Main causes of error are (i) the uncalibrated fluorosceence yield of the Ce:YAG screen and (ii) the different angular distribution of the fundamental and the harmonics.

15. Since the transmission of the beamline is almost constant over the 30-130 eV photon energy range, only the total FEL intensity is reduced between the spectrometer and the end station located about 25 and 70 m behind the undulator, respectively.