Metrology for X-ray Lasers

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Since the early days of research with synchrotron radiation [1], the respective accelerator-based radiation sources have constantly been further developed [2]. In the so-called *third generation*, the operation of insertion devices in the straight sections of storage rings has been optimized since the 1990s, especially the operation of undulators which emit highly brilliant radiation of variable polarization [3–6]. The latest developments are aimed at generating radiation pulses of less than 1 ps duration by means of optimized accelerator structures, e.g. with free-electron lasers (FELs) for time-resolved experiments on very rapid chemical processes. Among the FEL facilities which are currently in operation – FLASH in Hamburg [7], FERMI in Trieste [8], LCLS in Stanford [9] as well as SCSS [10] and SACLA [11] in Japan – all, except FERMI, are based on the principle of *Self-Amplified Stimulated Emission* (SASE). Thereby, very short electron bunches with relativistic energies are generated in a linear accelerator where a clearly better electron bunch focusing and compression, compared to ring accelerators, is possible. Due to this, the undulator radiation generated in a downstream undulator is so brilliant that there is a significant electromagnetic feedback with the electrons. This leads to microstructuring of the electron bunches with the period of the radiation wavelength, and to a coherent emission of FEL radiation which is amplified by many magnitudes. Meanwhile, photon energies between 20 eV and 20 keV are attained for pulse energies up to several mJ. Pulse durations between 10 fs and 500 fs thus lead to a radiation power of more than 10 GW in a single pulse.

As in an FEL pulse of a duration of less than 1 ps, approximately as many X-ray photons are available as in the case of synchrotron radiation in one second, FEL experiments can be carried out in a pulse-resolved mode. However, the SASE principle implies strong pulse-to-pulse variations – especially of intensity or pulse energy – which calls for real-time photon diagnostics. PTB has accompanied the development and the characterization of the respective detection systems from the very beginning [12]. For the worldwide first SASE-FEL with a user operation – FLASH in Hamburg – so-called *gas monitor detectors* (GMDs) were designed in cooperation with the *Deutsches Elektronen-Synchrotron* (DESY) in Hamburg and the *Ioffe Institute* in St. Petersburg and installed at the laser output in front of the experimental area [13]. These are based on the photoionization of gases and the detection of the generated photoions and photoelectrons. In Figure 1, such a system is shown. At typical gas pressures in the range between 10^{-2} Pa and 10^{-3} Pa, the detector transmission of FEL radiation for

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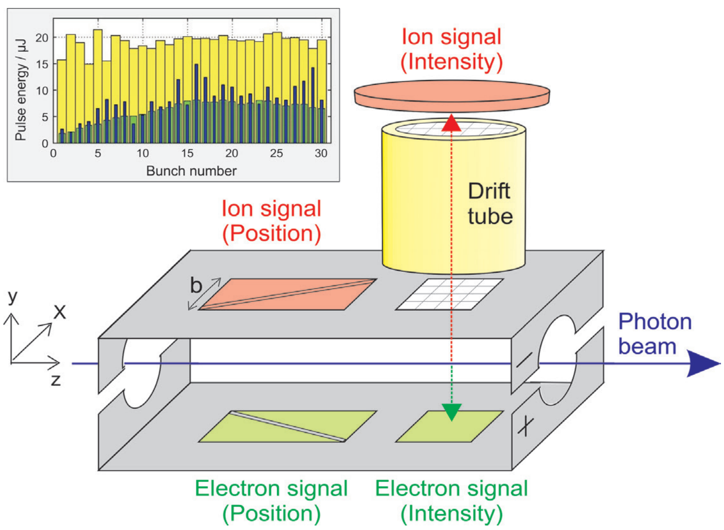

Figure 1:
Scheme of a gas monitor detector (GMD) at FLASH for the real-time measurement of FEL intensity and beam position. Top left, the display in the FLASH control system for pulse energies measured by means of a GMD is represented (blue: values for the current pulse train; yellow: maximum value of the pulse trains as of display start; green: mean values of the pulse trains as of display start) [13].

the experiments is beyond 99 %. Prior to their installation, the detectors were calibrated traceably in PTB's laboratory at BESSY II [14] – against calibrated photodiodes – to a cryogenic radiometer as a primary detector standard [15]. Therefore, they are continuously providing absolute information on pulse energies with relative uncertainties in the range of ±15 %. In addition, the FEL beam position can be measured by means of split electrodes [13].

Improved versions of GMD are also suited for mobile use, e.g. in order to quantitatively detect the radiation of *High Harmonic Generation* (HHG) [16], or for measurements carried out at other FELs [17–19]. For the harder X-ray region up to photon energies in the range of 20 keV, an option of signal amplification by the use of open multipliers was developed in order to compensate for the photoionization cross sections which are lower by orders of magnitude there.

An important aspect during measurements with DESY-PTB-GMDs on – meanwhile – all FELs in operation was the validation of the measurement and calibration procedure. Both at lower photon energies in the range between 20 eV and 25 eV [17] and in the X-ray range between 4 keV and 14 keV [18], radiometric comparisons took place at the Japanese FEL facilities SCSS and SACLA by means of a cryogenic radiometer of the Japanese metrology institute AIST/NMIJ. In these comparisons, a good agreement was achieved within the scope of the combined measurement uncertainties. Especially non-linear processes, such as multiphoton ionization, can influence the GMD measurements on an FEL essentially, compared to the calibration with synchrotron radiation, where the number of photons of a single FEL pulse was distributed to 10^8 to 10^9 pulses. Therefore, the non-linear influences on the photoionization were also systematically and quantitatively investigated at FLASH by means of focused FEL radiation [20–22]; however, these influences have proved to be negligible outside of focal regimes with irradiances below 10^{12} W/cm^2.

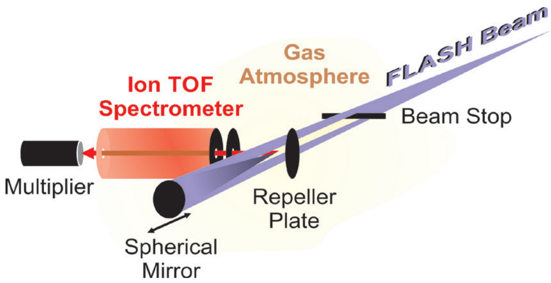

Figure 2:
Scheme of a gas-phase photoionization experiment at FLASH with an ion time-of-flight (TOF) spectrometer in the focus of a spherical multilayer mirror.

Figure 2 shows the experimental set-up of such a photoionization experiment, in which a spherical EUV multilayer mirror [23] in back reflection geometry generates a micro-focus. In the interaction zone of an ion time-of-flight spectrometer, irradiances of up to 10^{16} W/cm^2 could be generated at a photon energy of 93 eV, whereby the incident unfocussed radiation was blocked for the experiment. Mirror and focus could be displaced along the beam axis; the beam cross section and the irradiance were thus varied for the experiment in a defined way. Figure 3 shows a series of ion time-of-flight spectra for xenon [22], from which the respective distribution of the charge states generated during the photoionization can be read. Whereas the spectrum recorded at $2.5 \cdot 10^{12}$ W/ m^2, having the charge states Xe^{1+}, Xe^{2+} and Xe^{3+}, corresponds largely to the single photoionization of xenon from the 5*p*- or 4*d*-shell with subsequent Auger decay, the higher charge states at higher irradiances are the result of non-linear multiphoton processes. In the past few years, a controversial debate has been triggered on the ionization mechanisms, especially for the very high charges up to Xe^{+21} [22, 24–29], and the xenon work became the starting point for numerous further studies, also PTB's own studies in cooperation with various partners. The latter refer to comparisons between different noble gases [25, 30] with electron spectroscopy [31] as well as to measurements at different FEL pulse durations [29, 32].

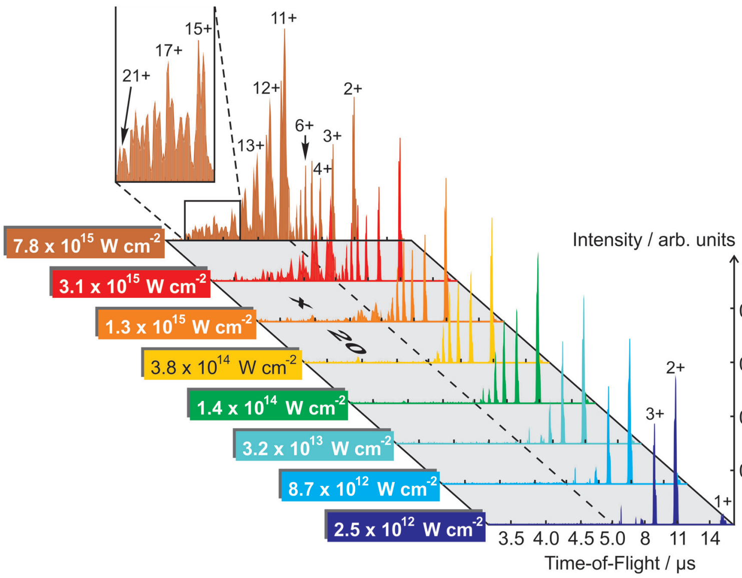

Figure 3:
Ion time-of-flight spectra of xenon, recorded at a photon energy of 93 eV and various irradiances [22].

Non-linear processes on the photoionization of gases were also used for additional FEL photon diagnostics. To obtain information, e.g., on the size of micro-focuses, the effect that an individual FEL pulse can depopulate a significant part of the targets in the focal zone by means of photoionization, was exploited. In this way, even for single photon processes, the ionization signal does not increase linearly to the pulse intensity any longer, but reaches saturation, from which the FEL beam cross section can be evaluated [33]. However, the two-photon double ionization of helium is – per se – a non-linear process which quadratically increases along with the irradiance [21, 26], from which the FEL pulse duration can be determined [34, 35].

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