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High repetition HHG source of ultrafast XUV pulses for the SXP instrument at the European XFEL

P. Grychtol[1], V. Vardanyan[1], D. Doblas-Jimenez[1], M. Heber[1,2], P. Bhardwaj[1], E. Tikhodeeva[1], S. Molodtsov[1,3] and M. Izquierdo[1]

[1]European XFEL, Holzkoppel 4, 22869 Schenefeld, Germany
[2]Deutsches Elektronen-Synchrotron DESY, Notkestraße 85, 22607 Hamburg, Germany
[3] TU Bergakademie Freiberg, Leipziger Strasse 23, 09599 Freiberg, Germany

E-mail: `grychtol@xfel.eu`

Abstract. This contribution presents the concept of installing a table-top high harmonic generation (HHG) source with high repetition rates at the Soft X-ray Port (SXP) instrument of the European XFEL. The compact implementation will enable studies in the extreme ultraviolet (XUV) range between 20 eV and 72 eV at the SXP interaction point using the existing experimental setup: a time-of-flight momentum microscope. This will allow for time-resolved investigations, which are complementary to those conducted with the soft X-ray SASE 3 undulator covering a spectral range from about 0.3 keV to 3 keV. This project will enable unique optical pump - XUV probe experiments, facilitate the preparation of soft X-ray beamtimes, contribute to the development of a robust research program at SXP, and provide opportunities for other groups at the European XFEL as well as the international user community alike.

1. Motivation

The exploration of ultrafast dynamics in condensed matter has been revolutionized by the advent of high-repetition-rate extreme ultraviolet (XUV) and soft X-ray (SXR) light sources combined with advanced photo-electron spectroscopy techniques. Free Electron Laser (FEL) and High-Harmonic Generation (HHG) sources, operating at megahertz (MHz) repetition rates, have enabled researchers to probe electronic core-levels and band structures, including non-equilibrium states with unprecedented temporal and momentum resolution. The integration of such advanced radiation sources with momentum microscopy, as demonstrated in recent studies, has opened new avenues for capturing the entire electronic structure of materials, thereby providing critical insights into ultrafast processes at atomic scales [1].

At the European X-ray Free Electron Laser (EuXFEL) [2, 3], the Soft X-ray Port (SXP) instrument represents a key innovation within this research landscape. Positioned at the SASE 3 undulator, the SXP instrument extends the capabilities of the EuXFEL by enabling femtosecond time-resolved X-ray photo-electron spectroscopy (TR-XPES) across a wide photon energy range from approximately 0.3 keV to 3 keV. This instrument is primarily designed for detailed studies of the electronic, spin, chemical, and structural properties of materials at surfaces as well as interfaces in general, leveraging the unique advantages of the EuXFEL, such as its ultra-high brightness, variable polarization, and tunable femtosecond SXR pulses [4].

The integration of a high-repetition-rate HHG source with a momentum microscope at the SXP instrument is a significant enhancement, allowing for simultaneous energy- and momentumresolved photo-emission mapping over the full Brillouin zone, in particular when the FEL is not available. This powerful combination facilitates in-depth exploration of ultrafast charge carrier dynamics and band structure evolution in complex material systems. The SXP instrument's design, including advanced X-ray optics and a time-of-flight momentum microscope, supports high-resolution studies that can push the boundaries of current materials science and solid-state physics research. The ability of the SXP instrument to perform complete TR-XPES studies across such a wide photon energy range, encompassing electronic, spin, and structural dynamics position it as one of the few global facilities capable of such comprehensive investigations [5]. In the following, this contribution outlines the concept of the high-repetition-rate HHG beamline to be integrated into the SXP instrument, specifically designed to operate in conjunction with the already installed and commissioned momentum microscope.

2. Overview of the compact HHG beamline

A top view of the SXP instrument model without the momentum microscope but including a section of the permanent beamline components and the outline of the FEL (blue) as well as HHG (pink) beam paths is presented in Fig 1. The FEL beam enters the SXP instrument on the right side of this figure and passes through the photon arrival time monitor (PAM) entering the laser in-coupling unit (LIN) thereafter - for details please check out [4, 6, 7] - leaving into the momentum microscope (not shown here) on the left side of the figure. While the PAM is supposed to monitor the relative arrival times between the SXR and synchronized optical laser (OL) pulses used to excite any sample under investigation, the LIN allows for coupling the OL beam and soon also the XUV beam into the path of the XFEL in a co-linear manner.

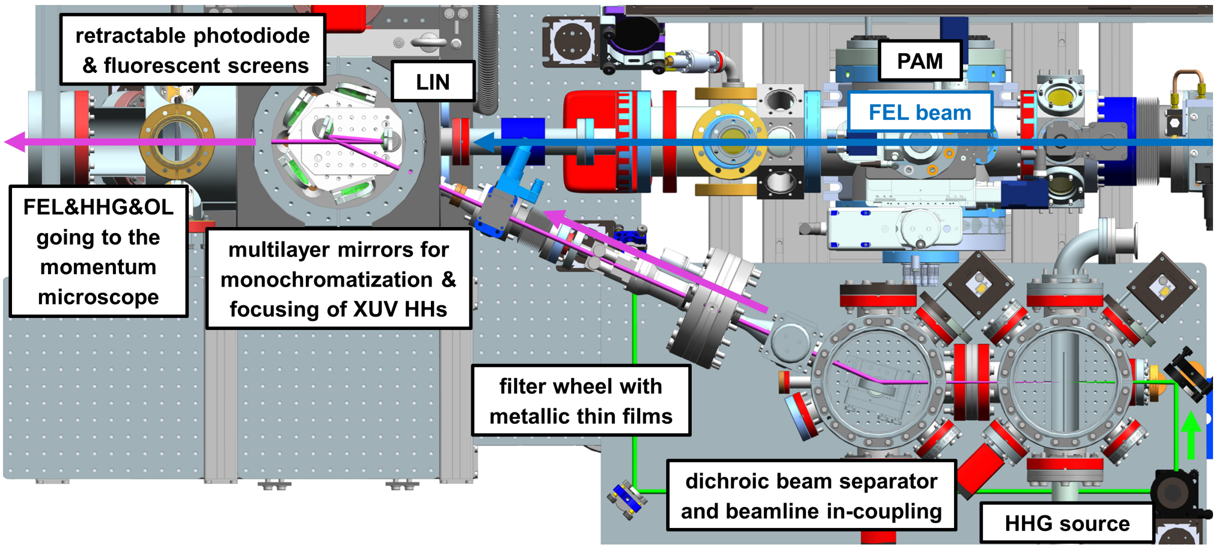

Figure 1. Top view of the compact HHG setup embedded into the SXP beamline.

The XUV beam (pink) originates in the center of the HHG source chamber, which is going to be placed on top of the optical bread board attached to the PAM chamber, as seen in the bottom right of Fig 1. It is generated by a nonlinear interaction of a tightly focused femtosecond laser beam (green) with a noble gas that is injected via a thin steel pipe into the HHG chamber. Both beams propagate co-linearly into the attached separator chamber hosting a B_4C-coated fused silica mirror, in which the XUV beam is reflected into the LIN where a pair of multilayer mirrors pass it on to the sample interaction zone. The fundamental laser beam is transmitted by the dichroic mirror and dumped to a large extent thereafter, while its residual reflected smaller portion is completely absorbed by thin metallic films in the filter wheel not reaching the LIN.

3. Generation of ultrafast XUV pulses

Ultrafast, few-femtosecond XUV pulses can nowadays be routinely generated via laser-driven HHG in gaseous media. The non-linear nature of the generation process results in a spectral comb of odd-order harmonics of the fundamental laser frequency with photon energies between a few eV and keVs depending on the generation conditions, which are mainly based on the available driving laser [8]. The currently installed standalone Yb-doped fiber laser amplifier system at the SXP instrument delivers approximately 300 fs short and 150 µJ strong laser pulses at a repetition rate of 333 kHz in the near infrared (NIR) region, at a center wavelength of 1030 nm. Its pulse train can be fully synchronized to the European XFEL and an internal acoustic-optical modulator allows for mimicking the characteristic 10 Hz bunch pattern.

These pulses can be spectrally broadened by a Herriott-type multi pass cell (MPC) through self-phase modulation in two 1 mm thin silica plates and they are subsequently compressed with a bunch of chirped mirrors resulting in a pulse duration of about 30 fs with a pulse energy of nearly 120 µJ [9]. Based on these laser specifications in combination with a tight focusing geometry, it will be possible to generate coherent pulses with a duration of less than 10 fs in the photon energy range from 20 eV to 72 eV. This will not only allow for exploring the entire Brillouin zone of most materials with the momentum microscope, but also for addressing the M absorption edges of most $3d$ transition metals, including the ferromagnetic elements.

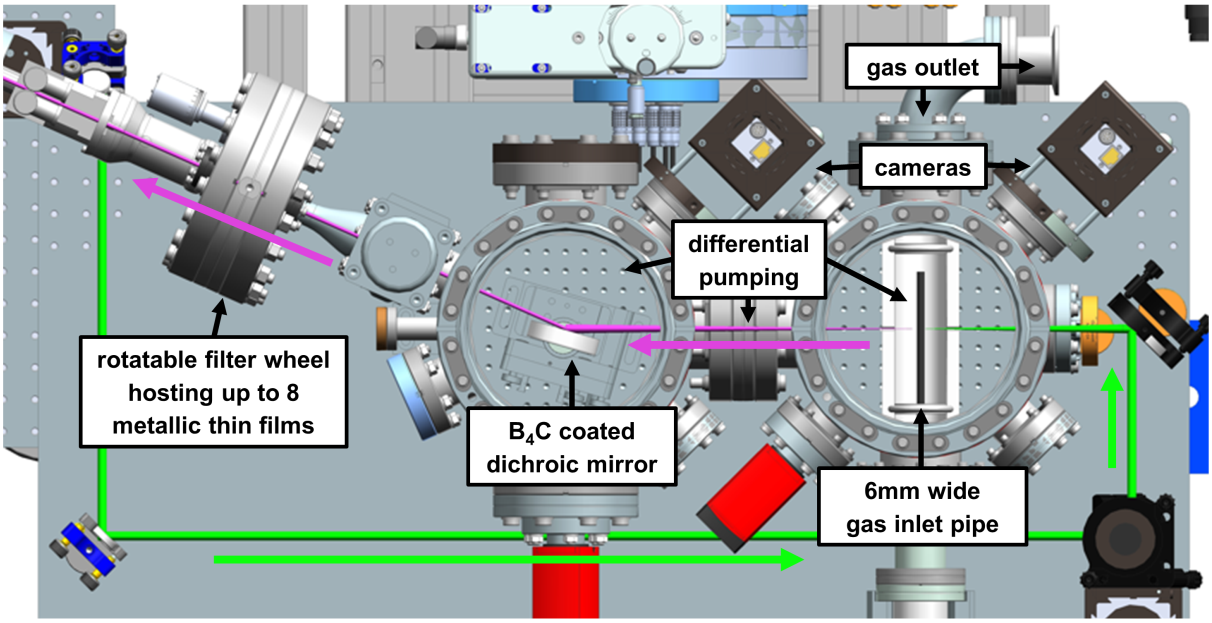

Figure 2. Top view of the HHG source and the separator chambers as well as the filter wheel.

For the actual HHG process, a lens with a focal length of 200 mm can be used, which is simulated to result in a beam waist radius of about 17 µm, when focusing the collimated 1030 nm laser with a beam waist radius of about 5 mm. Conservatively considering a pulse duration of about 35 fs and an available pulse energy of at least 100 µJ, a peak intensity of nearly $5 \times 10^{14}\,\mathrm{W/cm^2}$ can be obtained in the focus [10]. Thus, a cut-off photon energy of $3.17 \times U_P + I_P \approx 75\,\mathrm{eV}$ can be reached in Argon according to the single-atom classical model based on the Ponderomotive energy U_P and the gas-specific ionization energy I_P [11].

The Argon gas will be injected via a 6 mm thin steel pipe with an inner diameter of 4 mm, into which two holes will be drilled with the femtosecond driving laser beam at the beginning of the HHG process. For alignment purposes, the injection pipe is mounted to a X/Y/Z-manipulator and its position can be closely monitored with a dedicated camera system attached to the HHG source chamber. In order to minimize the reabsorption of the generated XUV in the conversion gas, an elaborate differential pumping scheme consisting of several apertures, powerful roughing and turbomolecular pumps mounted on top of the HHG source as well as the separator chambers will be implemented, as displayed in Fig 2.

4. XUV monochromatization and focusing

Once a bunch of XUV high harmonics is generated in the center of the source chamber, it propagates co-linearly with the fundamental NIR laser beam into the adjacent separator chamber via a 3 mm wide aperture drilled into the center of a flange connecting both chambers. It not only serves as a differential pumping stage, but it also blocks and absorbs a significant portion of the fundamental driving laser radiation due to its much higher divergence as compared to the XUV harmonics. In the following chamber, the XUV light is separated from the fundamental laser driver by a fully motorized B_4C-coated fused silica plate mounted close to the NIR Brewster's angle. At the chosen grazing angle of about 12°, the reflectivity of B_4C for p-polarized light at 1030 nm is about 3 %, while in the XUV range between 20 eV and 72 eV the reflectivity exceeds 70 % [12, 13]. This allows for a large portion of the generated harmonics to be reflected into the LIN and simultaneously transmitting as well as dumping the HHG driving laser to a large extent in the separator chamber.

The residual NIR light will be blocked by thin metallic films in the filter wheel mounted in between the separator chamber and the LIN hosting up to 8 pieces of various thicknesses and/or types. For example, a free standing 200 nm thin Al film suffices to completely suppress the 1030 nm light, while theoretically transmitting more than 60 % in a photon energy range from 20 eV to 72 eV [13]. Most likely the actual reflectivity and transmission of the individual dichroic components are somewhat lower due to thin oxidation layers, contamination and imperfections of the respective surfaces as well as interfaces. However, the calculated numbers are close enough to provide a reasonable estimate of the XUV flux to be expected in the sample region, proving that it is high enough to perform TR-XPES experiments using a momentum microscope in the designed configuration, as outlined in the following.

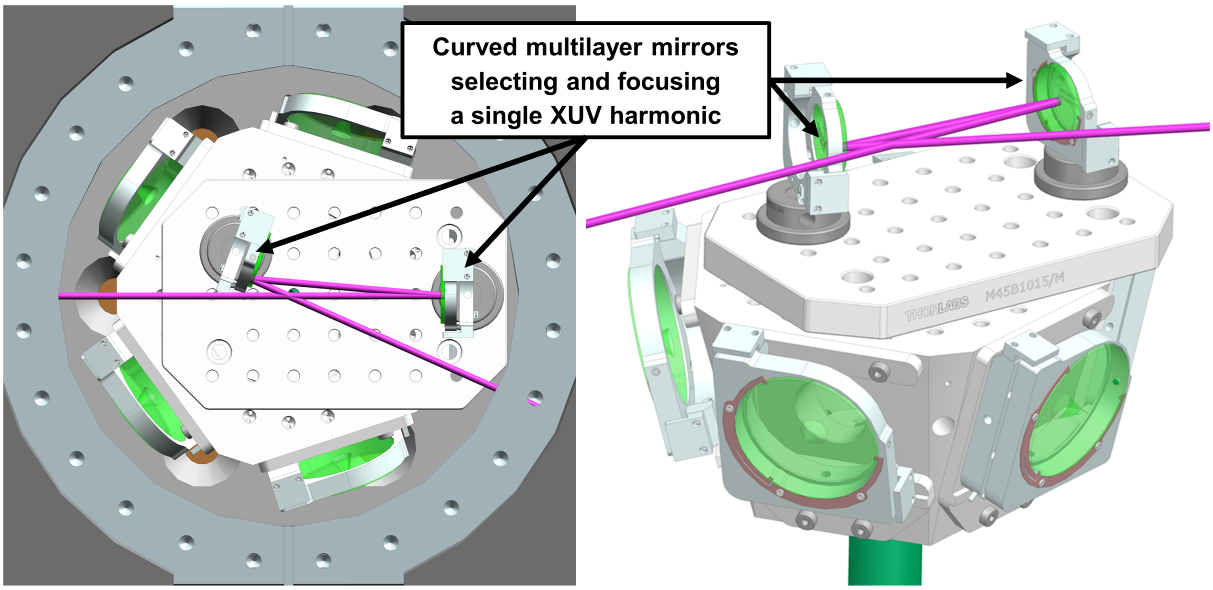

Figure 3. Top and side view of the multilayer mirror pair selecting and focusing a harmonic.

In order to prevent the photo-emission spectra to overlap as a result of the excitation from adjacent XUV harmonics, it is of utmost importance to select a single one to be focused on to the sample. As presented in Fig. 3, this is supposed to be accomplished using a pair of curved motorized multilayer mirrors placed into the LIN, which has the advantage over conventional grating techniques to conserve the pulse duration and chirp in the reflection process of the ultrafast harmonics. Beam propagation simulations suggest that by using a combination of convex and concave spherical mirrors, it is possible to image the HHG source point assumed to have a beam waist radius of 10 µm on to an approximately 2.5 m distant, nearly round spot in the sample region obtaining a beam waist radius of about 3 µm by carefully adjusting the incidence angles in a range of 5° to 8° [14]. Accordingly, the spherical mirrors will be commercially coated with a multilayer, such as Mo/Si or Al/Zr, to specifically select a single harmonic in the generated photon energy range known to provide a reflectivity of more than 30 % per mirror. Thus, it can be estimated that the total transmission of the beamline amounts to about 3 %. Assuming a HHG conversion efficiency at the order of 10^{-6} [11], an average power of a few µW can consequently reach the sample translating to about 9×10^5 photons per pulse. This is more than enough to photo-emit at least 1 electron per pulse, which is required to efficiently operate the delay line detector of the time-of-flight momentum microscope at the SXP instrument.

5. Summary

This contribution presents the development and implementation of a high-repetition-rate high harmonic generation (HHG) source of ultrafast extreme ultraviolet (XUV) pulses at the Soft X-ray Port (SXP) instrument of the European XFEL. The HHG source is designed to operate in conjunction with the existing laser infrastructure, providing ultrafast XUV light in the range of 20 eV to 72 eV, which complements the soft X-ray beam generated by the SASE 3 undulator. Thus, it will be possible to conduct a variety of ultrafast optical pump - XUV probe experiments, especially when the FEL is not available, and facilitate research into ultrafast dynamics in materials at the SXP instrument, particularly using the time-of-flight momentum microscope. This paper outlines the technical details of the HHG setup to be integrated into the beamline at SXP, including the laser-driven generation of XUV pulses and their propagation to the sample, and briefly discusses the potential of the beamline to advance materials science research.

6. Outlook

While the implementation of the presented HHG beamline will increase the scope of SXP, it holds significant potential to advance its research capabilities even further. For example, it is well known that the HHG spectrum can easily be adjusted from discrete peaks to a continuum spectrum by changing the pulse duration of the driving laser from a multi-cycle to a few-cycle pulse [8]. By this means it would be possible to reduce the XUV pulse length from the typical few fs region down to the attosecond regime. In addition, when adjusting the wavelength of the driving laser to longer wavelengths, it is feasible to increase the HHG cut-off energy up to several hundreds of eVs. This would allow for closing the energetic gap to the SASE 3 undulator.

Furthermore, it is important to note that all driving lasers at SXP can be fully synchronized to the European XFEL, which will not only enable to perform ultrafast pump-probe experiments combining the FEL with the HHG source, but also allow for time-resolved experiment using the HHG XUV source in conjunction with the dedicated SASE 3 pump-probe laser. The latter can also be used to generate femtosecond pump pulses in the wavelength range from about 250 nm to 15 µm when combined with an optical parametric amplifier [15]. Thus, the combination of ultrafast optical pulses, HHG and FEL pulses at the instrument will make SXP a unique and versatile instrument enabling multicolor experiments on the same sample. In particular, the ramification towards attosecond science aligns perfectly with the facility development plans.

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