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OPPORTUNITIES FOR TWO-COLOR EXPERIMENTS AT THE SASE3 UNDULATOR LINE OF THE EUROPEAN XFEL

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Abstract

As is well-known, the installation of a simple magnetic chicane in the baseline undulator of an XFEL allows for producing two-color FEL pulses. In this work we discuss the possibility of applying this simple and cost-effective method at the SASE3 soft X-ray beamline of the European XFEL. We consider upgrades of this method that include the further installation of a mirror chicane. We also discuss the scientific interest of this upgrade for the Small Quantum Systems (SQS) instrument, in connection with the high-repetition rate of the European XFEL, and we provide start-to-end simulations up to the radiation focus on the sample, proving the feasibility of our concept. Our proposed setup has been recently funded by the Finnish Research Infrastructure (FIRI) and will be built at SASE3 in 2020-2021. Detailed information is available at [1].

METHOD

The simplest way currently available to enable the generation of two closely separated (on the order of 50 fs) pulses of different wavelengths (which will later result in the two-colors) at X-ray Free-Electron lasers consists of inserting a magnetic chicane between two undulator parts as suggested in [2] and experimentally proven in [3, 4]. The scheme is illustrated in Figure 1-a. We propose to split the baseline SASE3 soft X-ray undulator into two parts with a magnetic chicane. Both parts act as independent undulators and will be referred further as $U1$ and $U2$. The nominal electron beam enters the first undulator $U1$, tuned to the resonant wavelength λ_1. After passing through $U1$, both electron beam and emitted Self-Amplified Spontaneous Emission (SASE) radiation enter the chicane. This magnetic chicane has two functions: first, it introduces a suitable delay between the electron beam and the radiation generated in $U1$. Delays from zero[1] up to the picosecond level can be obtained with a compact magnetic chicane of several meters length. Second, due to dispersion, the passage of the electron beam through the magnetic chicane smears out the microbunching at wavelength λ_1. As a result, when the -after the magnetic chicane- delayed electron beam enters the second undulator $U2$, the SASE process starts from shot-noise again. Therefore, if the undulator $U2$ is tuned to the resonant wavelength λ_2, then at the undulator exit one obtains a first radiation pulse at wavelength λ_1 followed by a second one with wavelength λ_2 delayed by a time interval that can be varied by changing the strength of the chicane magnets.

One must ensure that the electron beam quality at the entrance of the second undulator $U2$ is still good enough to sustain the FEL process. This poses limits on the maximum total pulse energy that can be extracted from $U1$ and $U2$. In particular, the amplification process in $U1$ should not reach saturation. Optimization of the maximum pulse energy also poses limits on the wavelengths choices The wavelength separation between the two pulses can theoretically span across the entire range made available by the undulator system, in the case of SASE3 between about 250 eV and 3000 eV. However, the impact of the FEL process on the electron beam quality depends on the radiation wavelength. Therefore, in order to maximize the combined radiation pulse energy that can be extracted, especially at large wavelength separations, the first pulse to be produced should be at the shortest wavelength. Moreover, the magnetic chicane strength should be large enough to smear out the microbunching at λ_1, unless the separation between λ_1 and λ_2 is larger than the FEL bandwidth.

An easy way to increase the flexibility of the scheme is to introduce a compact optical delay line to have full control on the relative temporal separation between the two pulses as shown in Figure 1-(bottom). Since the photon beam transverse size at the position of the magnetic chicane is, roughly speaking, as small as the electron beam, i.e. a few tens of microns, the length of each mirror can be as short as several centimeters. In order to simplify the design of the mirror delay line, one may fix the optical delay to a few hundred femtoseconds, thus avoiding the use of moving mirrors, and subsequently tune the delay by changing the current in the magnetic chicane coils. Therefore, the introduction of an optical delay line would allow one to sweep between negative and positive delays at a cost of a smaller delay tuneability, caused by a lower limit of the chicane magnetic field (while the optical delay is inserted).

Even the simplest way of generating two-color pulses at the SASE3 beamline of the European XFEL, in combination with the high-repetition rate capabilities of the facility is expected to enable novel exciting science at the two soft X-ray instruments: Small Quantum Systems (SQS) [5] and Spectroscopy & Coherent Scattering (SCS) [6].

Here we limit ourselves to the analysis of one science case for the SQS instrument.

SQS SCIENCE CASE

The two-color operation mode enables a large number of scientific applications based on a pump-probe excitation

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[1] In our case, due to radiation slippage in the subsequent undulators, the effective minimum delay between the two pulses of different colors is of the order of several femtoseconds.

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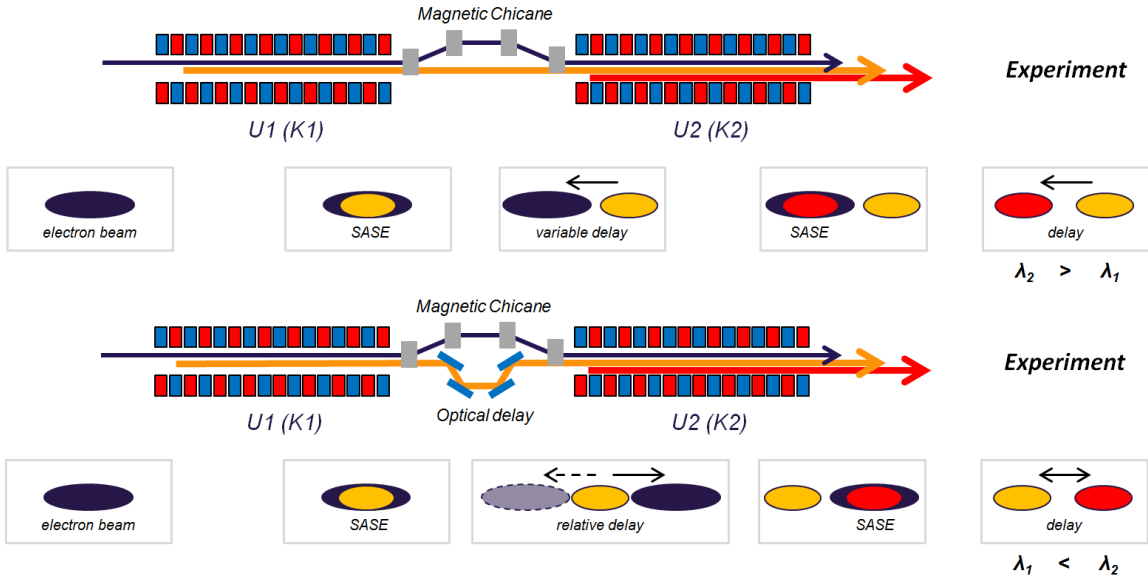

Figure 1: Schematic illustration of a simple two-color FEL technique without (top) and with (bottom) the addition of a compact optical delay line.

scheme with two individually controllable X-ray pulses. In the following a concrete example is discussed, which will make use of site-specific excitation in molecules possible at SQS scientific instrument of the European XFEL [5]. The excitation of a specific atomic site is enabled using soft X-ray pulses, since the radiation efficiently couples to the strongly bound core electrons, which are localized at the atomic site. Tuning the wavelength of the pump pulse to a specific threshold, a molecule can be excited at a well-defined atomic position. Using then the probe at another wavelength, which is connected to another core hole excitation, possible changes induced by the first pulse at a different site in the molecule are measured. Finally, the variation of the time delay between the two pulses provide access to the dynamics of this process, i.e. on the time, which is necessary to transport the information from one position in the molecule to another.

As illustrating example, we discuss charge transfer processes in a linear molecule, such as $I - C_n - H_{2n} - Cl$, composed of long carbon chain with two different halogen atoms, e.g. iodine and chlorine, at both ends. In the wavelength range accessible with the SASE3 undulator the $2p$ core electron of chlorine (threshold at 210 eV) as well as the $3d$ core electron of iodine (threshold at 630 eV) can be ionized. Considering a first pulse (pump) at a photon energy of 250 eV, the perturbation introduced by the XUV photon will be localized at the chlorine site, since core electrons of the other atoms (I and C) are still not in reach and cross section for valence ionization is weak. In the same way, choosing for the probe pulse the second photon energy at 630 eV assures that preferentially (i.e. most efficiently) the $3d$ electron at the iodine site is excited or ionized.

An efficient and informative experimental method to monitor the intramolecular processes is given by high-resolution Auger spectroscopy, ideally in combination with ion spectroscopy performed in a coincidence arrangement. The $3d$ Auger spectrum of iodine is located in the kinetic energy range around 400-500 eV arising mainly from the most prominent transitions to doubly charged states with electron configurations $4d^{-2}$ and $4d^{-1}4p^{-1}$ [7]. These lines are well separated from the corresponding Cl $2p$ Auger spectrum at kinetic energies between 165 and 175 eV [8] and other ionization processes taking place at the photon energies considered here.

Due to the high intensity of the FEL pulses sequential ionization processes are possible and likely to happen. As a consequence, the electron spectrum of the neutral parent molecule will be overlaid with emission lines arising from the ionization of the ionic species and of the dissociation fragments. In order to separate the emission from different species coincidences between electrons and ionic fragment can be used for a more detailed analysis. In fact, coincidence experiments will be one of the major experimental tools available at the SQS instrument, and are feasible due to the high number of X-ray pulses (up to 27 000 per second) at the European XFEL. This high repetition rate allows one to record data of high statistics for coincident measurements between electrons and ionic fragments coming unambiguously from the same molecule.

In a typical experimental scenario, first the Auger spectra would be recorded at the individual wavelength 250 eV and 630 eV, respectively, in order to obtain the one-photon reference spectra. In addition, electron-ion coincidence will provide charge and fragment resolved electron spectra would at these photon energies. In a second step, the Iodine $3d$ Auger spectrum – caused by the 630 eV photon pulse – will be monitored in the presence of the additional the 250 eV pulse. When the 250 eV pulse comes after the 630 eV pulse, the spectrum will be unchanged compared to the single color spectrum. When both pulses are overlapping or the 250 eV

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comes earlier, the observation of the iodine Auger spectrum for different delays between both pulses provide the information about the intermolecular processes. Changes of the kinetic energy position and of the intensity distribution within the $I3d$ Auger spectrum are the monitor to follow charge migration processes inside the molecule, i.e. to determine e.g. the time required to transmit the information about the creation of a $2p$ core hole on the chlorine site to the iodine atom. For small molecules this time scale is in the order to a few femtoseconds [9], so probably difficult to access with pulses of about 2 fs duration each. For longer carbon chains the time scale is expected to increase to about 10 fs or more and therefore well suited to be studied with the set-up at the SQS instrument.

Furthermore, by selecting in coincidence mode a fragment containing the iodine atom or the iodine atom itself, also information on charge transfer processes is made available. Compared to earlier work using an optical laser to initiate the fragmentation [10], the pump can be used in a very selective way, changing for example between excitations of the chlorine and the carbon atom. In this way a more versatile and detailed analysis of the complex intra-molecular interaction and on the related electron and nuclear dynamics will become possible.

SIMULATIONS FOR THE SQS SCIENCE CASE

For the particular science case at the SQS instrument discussed in the previous section, two fs-order-long X-ray pulses with a tunable relative delay are required. We considered a simulation scenario where a magnetic chicane and an optical delay line are installed at SASE3, see Figure 1.

Start-to-end simulations for the electron beam through the European XFEL linac to the entrance of the SASE3 beamline based on [11] were considered. The electron beam obtained is sent through the first part of the SASE3 undulator composed of 7 segments, with the first two segments not contributing to the SASE process and the rest 5 segments tuned at 630 eV. The photon beam then passes through the fixed optical delay line, while the electron beam goes through the magnetic chicane. The delay in the magnetic chicane can be adjusted with sub-fs accuracy, and can be set to under- or over-compensate the optical delay. We dump the numerically simulated electron beam distribution at the end of undulator $U1$ and use it for the FEL simulations in $U2$, which consists of 14 segments, with the first seven switched off and the remaining seven lasing at 250 eV.

The final step consists in performing wavefront propagation simulations thorough the SQS beamline, including a pair of offset mirrors and KB mirrors up to the sample position. After passing through the entire beamline, the photon beam can be focused at the sample position. One issue concerning the optimal focusing is related to the presence of the two separate sources for the two pulses. If we decide to tune the KB mirrors to image one of the two sources at a certain location, the image of the other will not only appear shifted in space, but will also be a subject to astigmatism. It is also possible to select an intermediate, imaginary source position between $S1$ and $S2$, which we call $S1.5$, and image it at the sample by properly tuning the KB mirror system. In this case, the images of $S1$ and $S2$ appear at positions denoted as respectively denoted as $I1$ and $I2$, while the image of $S1.5$ appears at location $I1.5$, where the sample is assumed to be introduced. This yields a good compromise in terms of beam sizes at the sample, Figure 2. Based on our simulations, the resulting photon fluxes would be sufficient to conduct the experiment proposed above.

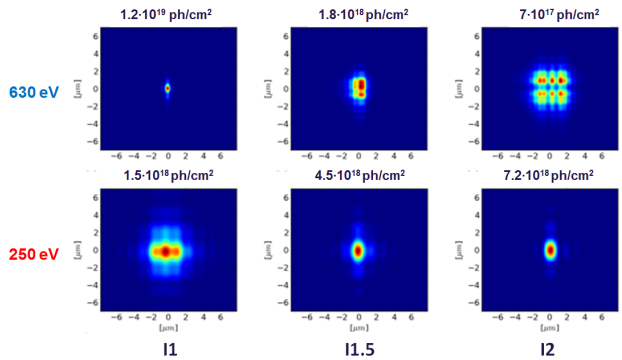

Figure 2: Radiation intensity distribution of both radiation pulses with different photon energies at various image planes. Peak photon density is provided above the plots. The method of an intermediate source reimaging would allow one to obtain comparable radiation distribution size as well as the photon flux.

While simulations presented in this paper were performed only for the purpose of illustrating the capabilities and the flexibility of the proposed setup, the same computational techniques produce results that may serve as a starting point for detailed simulation of the interaction between radiation and matter, and can be used to define and prepare experiments in great detail. Detailed information is available at [1].

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