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Investigating ultrafast structural dynamics using high repetition rate x-ray FEL radiation at European XFEL

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Abstract European XFEL is an international facility providing hard and soft x-ray free-electron laser radiation for user experiments with a wide range of scientific applications. Its superconducting linear accelerator enables high repetition rate experiments with a broad range of x-ray pulse delivery patterns. The combination of time-resolved experiments, providing access to the time-domain from sub-femtoseconds to milliseconds, with atomic resolution x-ray geometric and electronic structure determination methods is responsible for the bulk of scientific applications of European XFEL. In addition, the extreme x-ray intensities and coherence properties open new methods for studying matter out of equilibrium. After start of operation in 2017, the facility now harvests scientific applications with impact to the challenge areas climate and energy, health, environment and sustainability, and digitalization. Extensions of European XFEL aim to increase performance and capabilities for new scientific applications. An upgrade of the facility in the early 2030s will increase the applicability of European XFEL to solid materials and provide dedicated instruments for improved conditions in specific research fields.

1 Introduction

Recently established X-ray free-electron lasers (FEL) employ low emittance and high peak current electron accelerators to generate extremely intense, ultrashort duration and highly coherent x-ray radiation. Using synchrotron radiation as a seed and amplifying the radiation using the 'FEL principle' [1, 2] these sources provide x-ray radiation with properties complementary to x-ray synchrotron radiation from storage rings [3]. These properties make x-ray FEL radiation the superior source, e.g. for experiments investigating structural dynamics from the sub-femtosecond up to the micro- or even millisecond regime. These experiments allow advancing our knowledge about dynamic phenomena in complex solids [4], real chemical environments [5] and hard materials [6], just the way synchrotron radiation allowed to advance the understanding of static structures. The high intensity of the x-ray FEL pulses offers new methods for exciting matter [7, 8] and ultrafast control [9] not accessible for less intense synchrotron radiation. In addition, FEL experiments allow to record meaningful data from single events with objects as small as a single molecule, nanoparticle or nanostructure, thereby exploring properties not accessible in large ensembles or crystals.

While initial x-ray FEL experiments have been performed at low repetition rates of few to 100 Hz, high repetition rate experiments at frequencies from 10^3 to 10^6 events per second allow extending FEL applications to new sample classes and methods with much smaller cross-sections. This new capability for hard x-ray FEL radiation is based on high repetition rate accelerators and corresponding instrumentation and offers unique opportunities in a wide range of scientific disciplines.

The European XFEL is an international user facility for applications of x-ray FEL radiation located in Hamburg and Schenefeld, Germany [10]. It was created to provide to an international scientific community the means for deepening the understanding of structural dynamics of a large class of sample systems, thus pushing forward the frontiers of scientific knowledge, opening new scientific avenues, and contributing to solve major societal challenges [11]. The governance of the facility is based on an intergovernmental convention amongst currently twelve shareholder countries and its operation is entrusted to the European X-Ray Free-Electron Laser Facility GmbH. DESY as an internationally re-known accelerator laboratory has been assigned the task to operate and further develop the electron accelerator.

The facility comprises a 17.5 GeV electron energy superconducting linear accelerator (sc-LINAC), an electron switchyard with two beamlines for up to five FEL sources, and an experiment hall for five beamlines and more than ten scientific instruments [12]. In a first installation, phase three FEL sources and six scientific instruments were realized (see Fig. 1). The FEL sources SASE1 and

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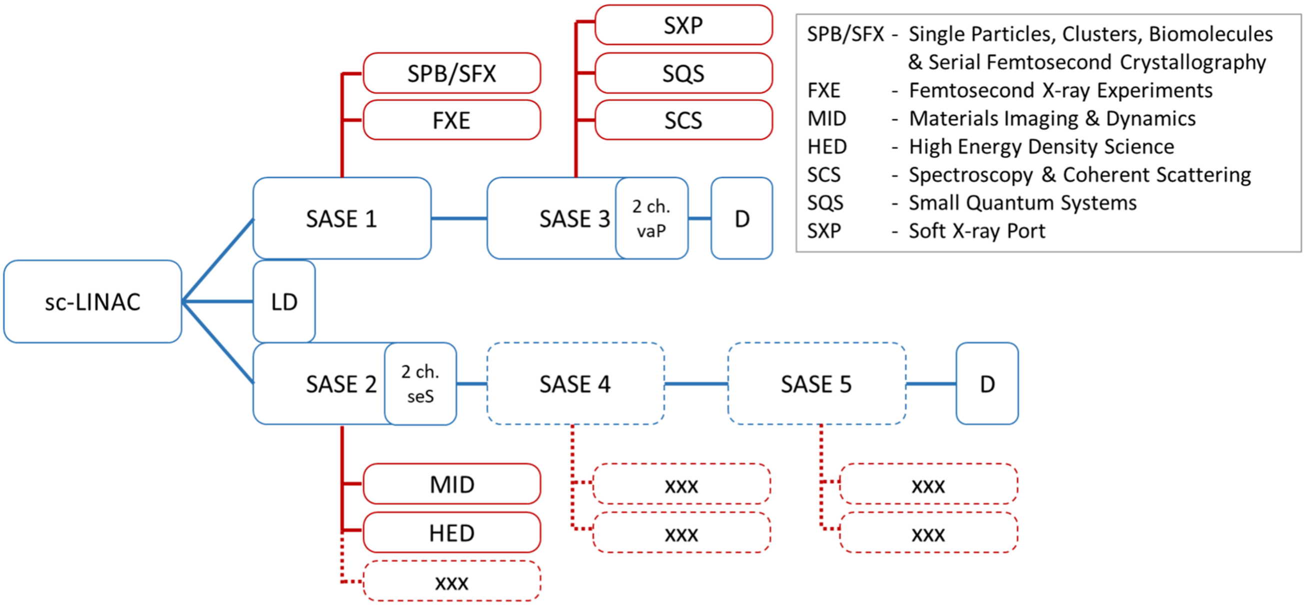


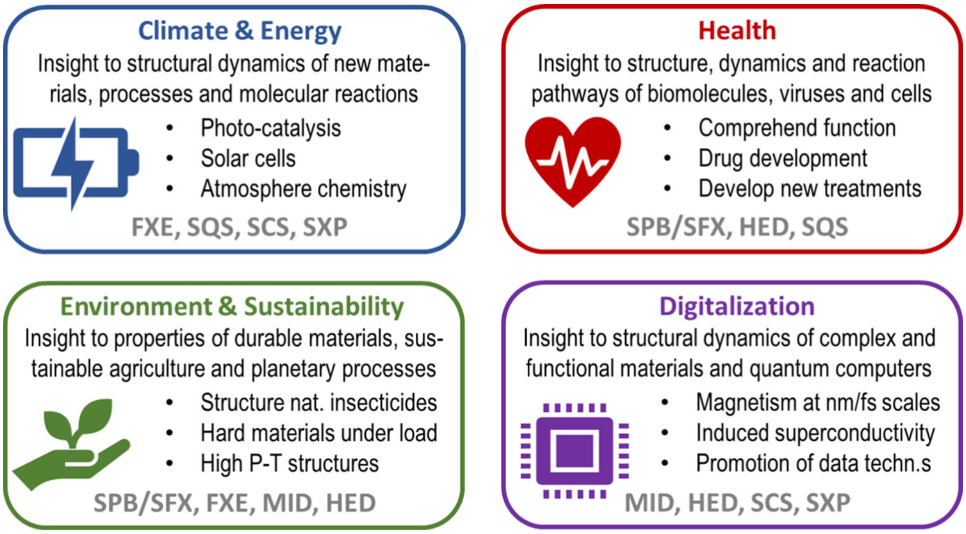
Fig. 1 Overview of the electron beam distribution (blue) and of scientific instruments (red) located at the various FEL sources, including their specialties (ch.—electron chicane; vaP—variable polarization; seS—Self-seeding; LD—LINAC dump; D—dump). Instrument full names are given in the insert. Broken lines indicate FEL sources and scientific instruments which could be added to the present facility

SASE2 cover the hard x-ray photon energy spectrum from about 5 keV up to presently around 20 keV. The scientific instruments SPB/SFX [13], FXE [14] (SASE1) and MID [15], HED [16] (SASE2) are located at these FEL sources. SASE2 has already been extended to include self-seeding and 2-color operation (see below). The third FEL provides soft and tender x-rays between about 0.28 and 3.0 keV photon energy and offers, in addition to SASE FEL radiation, a large number of special FEL operation modes. The scientific instruments SQS [17] and SCS [18] are located at this source, and with SXP a third SASE3 instrument is under construction. All scientific instruments operate more than one end-station, where each is optimized for a specific x-ray technique and/or sample environment, and are equipped with a high repetition rate laser system [19] and a laser-based timing synchronization system [20]. Through collaboration with the HiBEF consortium (see www.hibef.de) [16] the provision of lasers systems with several 100 TW pulse power [21] and 100 J pulse energy [22] to the user program of the scientific instrument HED is ensured. Very important for European XFEL had been its detector development program [23], delivering a total of six large area pixel detectors of three different types, each capable of frame rates up to 4.5 MHz, data rates of up to 14 GB/s/detector, and adjustable to the specific time-pattern of an experiment. Large amounts of scientific data, collected by means of these detectors, are stored centrally on currently 40 PB on high performance and approx. 100 PB on standard magnetic disc in order to be available to users for the scientific analysis. For its ambitious user program, European XFEL operates a user laboratory for the growth, selection and purification, sample delivery, and characterization of biochemical specimens [24]. In addition, a chemistry laboratory and various methods for sample characterization are available, as is an on-site fifty bed guest house.

The user facility employs quasi-simultaneous operation of all FEL sources where at a time one scientific instrument at each FEL source receives beam. Quasi-simultaneous refers to the fact that each electron bunch can be employed only once to generate high-quality FEL radiation since the FEL process degrades its energy spread. Accelerating in the sc-LINAC up to 27,000 electron bunches in 600 μs trains at a frequency of 10 Hz (so-called burst-mode), each bunch gets assigned to one of the FEL sources or a tune-up dump at the end of the LINAC. The x-ray FEL pulses are steered by means of mirrors to a specific scientific instrument and x-ray diagnostic data are collected for each pulse [25]. Within a pre-defined envelope, the users at these instruments can select the bunch pattern required for the respective activity of the on-going experiment. Operating 24 h and 6 days in approx. 30 weeks of x-ray delivery mode the facility near-term goal is to provide at the presently three FEL sources a total of 10.500 h for user experiments, and to increase this number to 12.000 h in the medium term.

2 Scientific applications and facility status

Following the initial proposal in 2002 [26], the construction of European XFEL started in 2009 with ground-breaking for the underground buildings. Important milestones of the facility construction have been the successful cool-down of the superconducting accelerator end of 2016, the first lasing at hard x-ray energies at SASE1 in May 2017, the start of user operation at two scientific instruments in September 2017 [3], and reaching the initial facility installation scope of the facility in May 2019 with the start of



Climate & Energy

Insight to structural dynamics of new materials, processes and molecular reactions

- Photo-catalysis
- Solar cells
- Atmosphere chemistry

FXE, SQS, SCS, SXP

Health

Insight to structure, dynamics and reaction pathways of biomolecules, viruses and cells

- Comprehend function
- Drug development
- Develop new treatments

SPB/SFX, HED, SQS

Environment & Sustainability

Insight to properties of durable materials, sustainable agriculture and planetary processes

- Structure nat. insecticides
- Hard materials under load
- High P-T structures

SPB/SFX, FXE, MID, HED

Digitalization

Insight to structural dynamics of complex and functional materials and quantum computers

- Magnetism at nm/fs scales
- Induced superconductivity
- Promotion of data techn.s

MID, HED, SCS, SXP

Fig. 2 Overview of the key societal challenges addressed by science experiments at European XFEL indicating which science instruments contribute most prominently to these areas

commissioning of the sixth scientific instrument HED. In 2022, the status of accelerator, FEL sources and instrument installations and operation is at a stage that one can consider the initial scope of the European XFEL facility to be complete and fully available to users and user operation. The now starting phase will see the harvest of scientific applications of the facility, a continuation to build up new capabilities, and a modest increase of capacity.

European XFEL has analyzed its contributions to the UN sustainable development goals [27] and similar lists, and has identified four societal challenge areas *Climate & Energy*, *Health*, *Environment & Sustainability*, and *Digitalization* as most relevant to the research performed at European XFEL (see Fig. 2 for details what is included under these topics). Experiments performed at European XFEL aim to increase fundamental knowledge relevant to these challenges. The results of these experiments provide detailed information about the structural dynamics of the atomic and electronics sub-systems of molecules and materials, crucial to further our understanding of processes relevant to develop solutions within each of these challenge areas. A specific advantage of European XFEL is the high repetition rate x-ray FEL pulses offering signal strength in combination with atomic specificity, sub-nanometer spatial resolution, and femtosecond time-resolution. Following three research examples of European XFEL are described.

Serial femtosecond x-ray crystallography (SFX) of ultra-small crystals makes unique usage of the intense femtosecond x-ray pulses focused in a few 100 nm beam at the SPB/SFX instrument [13]. The opportunity to collect single-shot diffraction patterns from individual crystals in a jet at up to 3.25 kHz average frame rate offers the opportunity to collect complete data sets within tens of minutes, also called MHz SFX. At the same time, the pulse and crystal resolved data frames allows avoiding the collection of data from radiation damaged crystals and allows one to distinguish between different crystal species or biomolecular structures—that is, to explore heterogenous populations. Adding laser excitation and varying the delay between an optical laser pump and an x-ray probe pulse one can investigate the evolution of the molecular structure with down to femtosecond resolution on femtosecond to millisecond time-scales. To probe bimolecular reactions on microsecond to millisecond, time-scales specialized jets are used to mix the molecule with a reactant, thus initiating a biochemical reaction which can be followed in time as the reaction proceeds. This capability of European XFEL offers unique access to problems directly relevant to the societal challenge areas *Health* and *Environment & Sustainability*. A recent experiment employing MHz SFX determined the Tpp49 Aa1 structure of a bacterial insecticide to a final resolution of 2.2 Å [28]. The nano-focus option at the SPB/SFX instrument, in combination with megahertz repetition rates, was used for rapid and high-quality data collection from natively grown nanocrystals paving the way for investigations of the structure and dynamics of bacterial insecticides. As a result of the study the insect line *C. tarsalis* was suggested as a new target organism for this class of insecticides and models could be identified for investigating the cellular pathways by which toxicity is elicited. These results will allow to shed further light on the mode of action of these toxins and SFX experiments [29] thus will aid the design of optimized and safe insecticides to combat mosquitoes as vectors of emerging diseases such as Dengue, West Nile Virus and Zika.

Ultrafast time-resolved soft and tender x-ray scattering offers unique capabilities to study the dynamics of complex and magnetic materials. These samples usually are prepared in the form of structured or disordered solids, or even come in the form of functional materials. The SCS instrument has been designed for studying ultrafast dynamics of complex solids using soft and tender x-rays employing diffraction, imaging, or by x-ray absorption and high-resolution inelastic x-ray spectroscopy [18]. The instrument allows to study both geometric structure as well as electronic structure with time-resolutions of a few femtosecond. By means of tuning the incident photon energy to the respective absorption edges of the atom under study, the before mentioned techniques can be performed resonantly. The photon energy range of the SASE3 FEL comprises from 0.5 to 1.6 keV the 3d transition metal $L_{2,3}$-, rare earth $M_{4,5}$- or actinide $N_{4,5}$ -edges, particularly relevant to complex and magnetic materials. This capability of European XFEL offers unique access to problems directly relevant to the societal challenge area *Digitalization*, which was demonstrated in a recent experiment

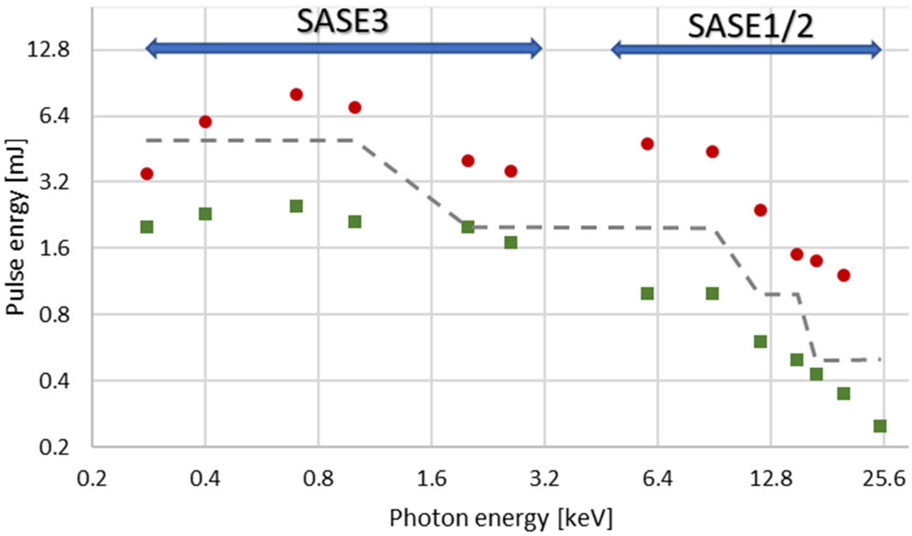


Fig. 3 Range of pulse energies attainable for a given photon energy, 250 pC bunch charge and optimized electron energies. Red circles and green squares correspond to maximal obtained, resp. calculated saturation values. The broken line indicates the expectation values used for preparation of experiments and the respective FEL sources are indicated

by observing the formation and dynamics of sub-10 nm magnetic structures in FePt nanoparticles [30]. Non-resonant tender x-ray diffraction was used to determine the formation of a spin-wave soliton and its spin precession frequency with a time resolution of 60 fs. Studies like this provide new and fundamental knowledge about materials response to writing, erasing and stability of magnetically recorded information, all of which are needed to develop new magnetic storage media having higher capacity, requiring less energy, being reliable and environmentally sustainable [31].

Femtosecond x-ray spectroscopy and scattering of photo-active solvated molecules contribute important information to the understanding of their photo-excitation and subsequent relaxation. The FXE scientific instrument combines for this purpose femtosecond laser and x-ray beams with liquid jets to perform both femtosecond time-resolved x-ray solution scattering using wide-angle X-ray scattering (WAXS) and x-ray spectroscopy. WAXS employs a large area pixel detector and x-ray spectroscopy various crystal analyzers and position sensitive pulse resolved detectors. While the time-resolved WAXS signal is sensitive to atomic structure, in particular to the distances between the atoms comprising both solute and solvant, the spectroscopy is sensitive to the occupation of specific electronic states, for example, the oxidation or spin states of the sample molecule. One particular strength of FXE is the ability to probe the electronic configuration of the valence states by very weak valence-to-core X-ray emission spectroscopic (XES) transitions as the sample undergoes photoreaction [32]. These signals are sensitive to the bonding electrons in molecules or the valence and conduction band electrons in crystalline materials making them a powerful tool to probe dynamical processes such as catalytically activated chemical reactions or charge carrier relaxation in functional materials. This capability of European XFEL offers unique access to problems directly relevant to the societal challenge area *Climate & Energy*. In a prototypical experiment, studying the excitation of solvated iron metallogrid complexes and using femtosecond time-resolved XES at the Fe $K_{\alpha1,2}$ and K_β lines it was possible to show that the lifetime of the metastable photo-induced spin state increases significantly from a few to over 100 ns for the tri-iron complex [33]. The use of XES was crucial to obtain this spin excitation lifetime information which is not accessible through optical spectroscopy. These results demonstrate the importance of ultrafast time-resolved x-ray techniques to further develop our understanding of photo-active molecules for their application as energy converters, catalysts or sensors [5].

The success in the above described scientific applications depended on the performance of FEL radiation and the beam modes provided by European XFEL. It is important to realize that European XFEL accelerator generates up to 600 ms long trains of electron bunches sent to all FEL sources at once. This parallel operation is today still unique amongst the FEL facilities. For the user experience, both single x-ray pulse performance and optimal conditions of multi-FEL operation are important. All performance goals for the individual FEL sources have been achieved, and efforts are under way to optimize pulse energies and duration. Other x-ray pulse properties are less frequently requested with properties outside the current conditions. Figure 3 shows attainable pulse energies as a function of photon energy, demonstrating the increase of pulse energies relative to saturation values by regularly operating in the over-saturation regime. Maximizing pulse energies and repetition rates enabled average x-ray beam powers above 30 W, both for hard and soft x-rays. This number already presents significant technological challenges, since the corresponding peak power during the bursts is approx. 200 times higher. Current operation uses 250 pC bunch charge and a compression leading to typical electron bunch durations of approx. 50 fs (fwhm) [3]. Ultrashort X-ray pulse durations and a high stability of x-ray arrival, each of order of 10 fs (fwhm) at a bunch charge of 250 pC [34], have been achieved, too, and will be important for scattering and spectroscopy experiments utilizing ultra-high time resolution. Successful campaigns to provide x-ray pulses with durations at the single femtosecond level and below have been performed using linear and non-linear compression of the electron bunch phase space, at times coupled with fresh-bunch lasing [35, 36].

Multi-FEL operation employs usually 2.25 MHz intra-train repetition rate, allowing up to 1300 electron bunches per train to be accelerated, of which typically up to 1100 can be distributed in a dedicated fashion to the three FEL sources. The bunch trains

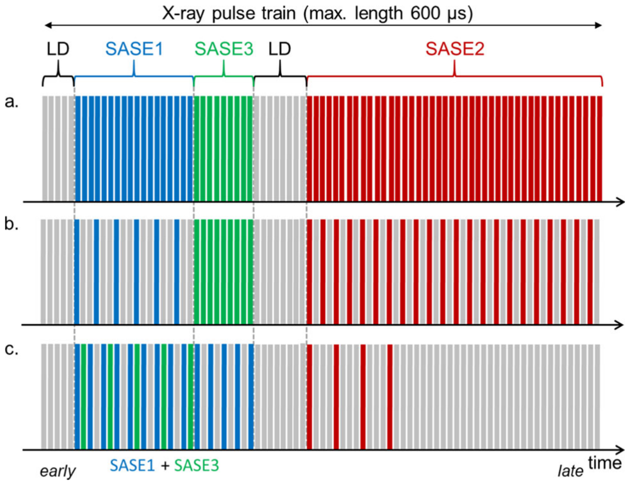


Fig. 4 Electron bunch patterns including FEL assignment [39]. **a** shows a pattern where the max. number of bunches is used for FEL generation, well separated for the three sources, **b** down-selection of pulses for SASE1 and SASE2 due to the experiment requirements, and c. interleaved delivery for the SASE1/3 FELs. Inter-bunch frequency (1.125, 2.25, 4.5 MHz) and the ratio between bunches going to SASE1/3 and SASE2 can be chosen according to experiment requirements

accelerated at 10 Hz in the main accelerator are normally distributed in a sequence “LD—SASE1 & SASE3—LD—SASE2” from head to tail of the train (compare Fig. 4). The portions send by fast kicker magnets to the LINAC dump are used for feedback and switching between electron beamlines. The ratio between SASE2 and SASE1 & SASE3 can be freely chosen. Differentiation of bunches assigned to SASE1 and SASE3 FELs is achieved using fast kickers producing betatron-oscillations around the straight orbit [37]. The kicked bunches do not lase in the subsequent undulator, but will still produce non-coherent synchrotron radiation that needs to be filtered out on its path to the sample. Different portions within an electron bunch train can receive different acceleration conditions as defined by the LLRF system [38], thus optimizing the delivery conditions for each of the FELs. For SASE1 and SASE3, located in the same electron beamline (see Fig. 1), the use of so-called interleaved delivery patterns enables to use longer pulse patterns (up to 500 μs) combined with smaller intra-train repetition rate (typically 1128 or 564 kHz and down to 112 kHz independently selectable for SASE1 and SASE3 experiments). In addition, each instrument can select according to the needs of the experiment between zero and a defined maximum of pulses for each delivery pattern. The extreme flexibility for the x-ray delivery pattern is a particular achievement of the accelerator and electron beam distribution systems, going well beyond the requirements defined by the experiments prior to start of operation. It requires that special attention goes into the observation of transients and the application of feedback schemes to ensure that the beam conditions remain stable independent of the delivery pattern, and in particular unchanged at the other FELs.

Various special FEL modes have been implemented to enable optimized conditions for specific applications and are at the disposal of the users. Most important is the self-seeding at the SASE2 FEL, allowing to provide radiation with much narrower spectral bandwidth, somewhat reduced pulse energies, but significantly increased spectral flux [40]. This mode is required in particular by experiments needing a FEL bandwidth significantly smaller than the natural FEL bandwidth, like inelastic scattering, nuclear resonance spectroscopy or x-ray photon correlation spectroscopy. The special realization at European XFEL employs two chicanes and crystals to circumvent the issue of x-ray beam power on the crystals, in particular relevant for the burst mode operation and at x-ray energies below 9 keV [41]. Another mode available to users at SASE2 and SASE3 is the provision of double pulses with different photon energy. This mode is most often requested by experiments using the first x-ray pulse to excite the sample (e.g. heating or electronic excitation), and the second pulse for probing the excited system. Electron chicanes can generate delays of few 100 fs at SASE1,2 and approx. 1 ps at SASE3. For SASE3 a fixed delay x-ray chicane to enable zero crossing is under construction. The pulse and photon energies of the two x-ray pulses are limited in case the ‘sequential 2-color mode’ is applied [36]. Additional schemes, where ‚fresh slices’ of the bunch are used in the two sub-sequent undulators, are under investigation and can avoid some of the limitations set by the sequential 2-color mode. Tested, but not yet available for users are operation modes to deliver 30 keV x-ray pulses, generated at SASE2 with pulse energies in the few 100 μJ regime, and fully variable polarization in the soft x-ray regime, provided by means of four APPLE-X segments behind the SASE3 FEL [42].

3 European XFEL extensions and upgrades

The most important improvement of the facility at present concerns the reduction of efforts for performing FEL experiments, both by users and staff. FEL experiments are complex due to the requirement to have at the same time x-ray beam, pump probe laser and sample delivery at a performance state allowing to take data. In addition, the feature to collect data for each single pulse leads to an enormous data acquisition effort, which has to run smoothly and produces huge data quantities to be managed, down-selected, calibrated and analyzed. An increased reliability of each of the sub-systems of the experiment, control systems enabling to easily monitor their performance, and automation software to ease adjustment and setting of parameters will be important ingredients to make experiments at European XFEL simpler and less resource demanding. In addition, software routines to support data selection, calibrations and standard data analysis will reduce the data quantity and increase the data quality per experiment considerably. These improvements will increase the experiment success rate and will further be important to enable access to European XFEL for research groups not expert in FEL techniques, limited in size, and rather interested in applying x-ray FELs to their samples and scientific problems.

In order to enable new scientific applications, requiring new capabilities of European XFEL, two extension projects are on-going and few major extensions are under preparation. On-going are the installation of additional undulator segments at SASE3 to provide variable polarization. This FEL property is strongly requested by experiment studying magnetic systems and chiral molecules. As well the construction and commissioning of the SXP scientific instrument has advanced and will provide from 2023, an open-port for soft x-ray applications not covered by the SCS and SQS instruments. An instrument for photo-electron spectroscopy has already been prepared.

Under preparation is three extension projects. The first concerns the possibility to provide high energy x-ray FEL radiation in the range 30–100 keV by the means of superconducting undulators (SCU) and harmonics generation. The provision of this very high photon energy FEL radiation would open a new field of FEL science applications. Expected areas of scientific use are experiments either studying very hard materials, for which the ability to transmit bulk specimen is crucial, or applying experimental techniques requiring higher photon energies to provide meaningful data, e.g. in terms of large enough momentum space coverage. The concept foresees to employ SCUs with a total magnetic length of approx. 24 m after the SASE2 FEL in order to generate x-ray FEL radiation above 30 keV. A prototype SCU with a period of 18 mm, and a magnetic field of 1.8 T has been designed and will be tested until 2025 [43]. A second extension is the provision of x-ray FEL radiation with attosecond pulse duration. Provision of x-ray pulse duration around and below one femtosecond will be of crucial importance to study changes of electron density after excitation and as a function of time, e.g. charge migration during a chemical reaction. For this extension, the electron bunch phase space will be manipulated by either passive beam structures [44] or intense optical laser pulses [45], energy modulation be converted into density modulations, and propagated either to the SASE1 or SASE3 FELs where they generate FEL pulses with sub-femtosecond duration. A corresponding project is currently started and an extension of this capability to SASE2 is possible at a small extra effort. A third project is the implementation of a prototype X-ray FEL oscillator in the SASE1 source [46]. This source would provide fully coherent and narrow-band x-ray pulses needed for non-linear and quantum optical applications, but may provide critical performance as well for high resolution x-ray spectroscopy experiments.

Another extension of the present facility will be the construction of additional FEL sources at the two vacant positions (compare Fig. 1), and the respective beamlines and scientific instruments. The sources will offer additional capacity and capabilities at European XFEL. The scientific areas and requirements to these additional instruments and FEL sources need to be defined within the next years, in order commissioning of them could start before the end of the decade.

Beyond these various extensions of the European XFEL scope, considered feasible for the remainder of the current decade, a real upgrade of the facility requires significant preparation to be executed during the first half of the 2030s. Such an upgrade will be important to create more and better matching capacity for scientific experiments, responding to the increasing needs for more sophisticated and specialized FEL instruments, but as well for an increase of users, respectively, a larger demand for experiment time. The upgrade will allow both, to increase the impact x-ray FEL science has to societal challenges and to enhance the methodological development of x-ray FEL applications. Upgrading European XFEL will build on the capabilities offered by its unique superconducting LINAC. Two major elements of such an upgrade are under consideration.

First, an upgrade of the superconducting accelerator offers a higher variability of pulse patterns delivered to the instruments. Such an extension is important to enable experiments needing a moderate high repetition rate of 100 to few 10.000 Hz, currently not available due to the 10 Hz burst-mode operation, but at the same time continuing to provide very high repetition rate pulse patterns, e.g. for experiments probing the evolution of the sample on nanosecond to microsecond time scale, and highest photon and pulse energies. As part of such an upgrade, an improved burst-mode might offer electron energies reaching up to 20 GeV combined a reduced emittance, longer pulse trains, tailored pulse properties, and highest photon energies. A second operation mode, called cw-mode, would make use of a new RF system allowing operation of the accelerator with an adjustable duty factor. Instead of the present 0.6% duty factor (burst-mode), approx. 15% with an electron energy around 12–14 GeV (long pulse mode) or 100% (continuous delivery) for 7–8 GeV have been proposed [47]. This mode offers the possibility to deliver significantly more x-ray pulses per second compared to the burst mode. For a 15% duty factor x-ray pulse delivery rates of 15.000–150.000 pulses/s could be obtained, using 100 kHz–1 MHz repetition rate within the pulse. The cw-mode, for which limitations due to absolute electron and x-ray beam powers still need to be defined, will be important for experiments requiring a combination of negligible sample excitation with highest repetition rates to collect enough signal.

The second upgrade element is the construction of a second electron beam distribution to feed a series of new and optimized FEL sources and a second experiment hall allowing to place more scientific instruments responding both to specific science requirements and an increasing demand for x-ray FEL time. Part of this upgrade will be to optimize the suite of all FEL sources and scientific instruments in a way that dedicated instruments using tailored x-ray FEL beams provide optimal conditions for specialized scientific application areas. The tentative time plan for preparing the upgrade foresees a preparation of scientific cases and technical designs before the end of the decade.

4 Perspectives of future scientific challenges to be addressed by European XFEL

Hard x-ray FEL experiments are presently converging from a phase of exploratory experiments, during which new methods and techniques were developed and tested on prototypical or highly standard sample systems, to a phase focused on targeted scientific applications and their requirements. Examples of such areas are SFX for drug discovery [48] or chemical crystallography [49], water research [50], or systematic studies of photo-catalysts [5]. These application areas are supported by several scientific instruments around the globe and all instruments at European XFEL assign significant fractions of their user time to them. Continuing the development of methods and sample environments will be important to bring these applications to full fruition.

New areas, which only start to be investigated, are the systematic space- and time-resolved understanding of hard materials and the study of electronic excitations and charge migration at the femtosecond level and below. The first domain will significantly benefit from the future availability of high energy x-ray FEL radiation, say 30–80 keV, with its ability to penetrate the bulk of hard materials [6]. Important applications address how these materials respond dynamically to load or external excitation and the investigation of stochastic processes such as crack propagation, defect formation, or nucleation. The second application area will require and exploit an improved time-resolution of single femtoseconds and below [51]. These studies will deepen our understanding about the interplay of the atomic and electronic systems in complex molecules and materials and the results will enable to design better excitation methods and molecules exhibiting optimized reaction pathways.

Describing the x-ray FEL experiment of the future one might best use the description of the molecular movie. Employing the unique properties of X-ray FELs to adjust spatial and temporal resolution, such movies will in future allow researchers to follow atomic motion and electronic excitations from the shortest (attosecond) time regime to the rearrangement of macroscopic numbers of atoms in the millisecond regime. Probed samples will range from small molecules, via complex bio-systems, to real materials, and investigations will be performed under operando conditions. Understanding and describing a probed volume at all length scales, from the atomic to the macroscopic regime, and being able to zoom in the relevant voxels will employ techniques developed at synchrotron sources today, and will imply a huge data management and software challenge. Materials research will benefit the most from these movies, since through these movies a profound understanding of the dynamic processes responsible for specific materials characteristics will develop. Such understanding will contribute to the development of new energy conversion materials, catalytic systems, and entire systems such as e.g. batteries.

European XFEL will pave the grounds for this transformation in the years to come through harvesting scientific applications using present-day capabilities. Extensions of the facility during the current decade and an upgrade in the early 2030s will ensure that the development of the capabilities and instrumentation keeps pace with the increasing demands of our society to provide scientific methods to address environmental and societal challenges.

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