FLASH and the FLASH2020+ project - current status and upgrades for the free-electron laser in Hamburg at DESY

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Abstract

The free-electron laser in Hamburg, FLASH, is the first extreme ultra-violet and soft X-ray free-electron laser (FEL) user facility and has been continuously upgraded since its start in 2005. Further major works are currently underway within the FLASH2020+ project that pioneeringly implements full repetition rate external seeding at a superconducting accelerator facility. With fully tunable undulators providing variable polarization FEL pulses, we expect FLASH to turn into the ideal spectroscopy machine for ultrafast processes within the coming years.

Keywords: free-electron laser, ultrafast dynamics, spectroscopy

1 Introduction to the facility

FLASH is a photon science user facility, operating two undulator lines in parallel and covering wavelengths from the extreme ultra-violet range down to the soft X-ray range. As single-pass FEL based on the principle of self-amplified spontaneous emission (SASE), it produces typical pulses of several ten to few hundreds of femtoseconds duration at single pulse energies of up to one millijoule. Driven by a superconducting accelerator, FLASH provides a high number of pulses per second (5000 or more). Electron bunches are produced

Fig. 1 Aerial view of the FLASH facility in the DESY campus. Electrons are generated in the hall on the top left and are fed into the grass-covered tunnel. At the bottom right, the Albert-Einstein experimental hall of FLASH1 and below the Kai-Siegbahn experimental hall of FLASH2 are visible. FLASH crosses the PETRA ring in between two experimental halls at the bottom center and on the top right.

ten times a second in bursts of typically up to about 500 pulses with 1 μs spacing [1].

The roots of FLASH lie in the TESLA project for a linear particle collider from the mid 1990s. For testing the new superconducting accelerator technology, a TESLA test facility (TTF) was conceptualized to also generate photon pulses in a single-pass SASE FEL scheme. In the late 1990s, TTF1 was realized [2] and early experiments using the photon pulses were performed at wavelengths in the range from 80 to 120 nm [3]. The second phase, TTF2 started operation in 2004 and was opened to photon science users in 2005. Renamed to 'FLASH', the facility received continuous upgrades. In this pioneering phase, a large set of fundamental method developments and experimental insights have been gained at FLASH that now form the basis for many of the experiments at other FEL facilities worldwide. The most recent major upgrade of the facility (completed in 2016), FLASH2, included the installation of a second electron beamline and a second experimental hall [4].

The undulator section at FLASH1 consists of fixed gap undulators, delivering horizontal FEL polarization and requiring changes in the electron energy in order to tune the delivered wavelength. A wavelength range between 51 nm and 4.2 nm was covered. Downstream of the FLASH1 main undulators, we operate an electromagnetic undulator with nine periods to produce synchronized radiation pulses in the THz spectral range from 1.3 to 30 THz and pulse energies up to about hundred μJ [5].

The undulators at FLASH2 provide horizontal polarization and a variable gap. This allows for easy tuning of the wavelength and at a given electron energy, the tuning range between shortest and longest wavelength is about a factor of three. Tuning undulators and electron energy allows FLASH2 to cover a wavelength range from 90 nm down to a bit below 4 nm. Substantial (μJ level) third harmonic radiation reaching beyond the $3d$ transition metal L-edge resonances has been observed and provided to users.

A third electron beam line is devoted to the FLASHforward experiment. Here, the FLASH electron bunches can be injected into a plasma cell, where the head of the electron bunch generates wakefields which either further accelerate parts of the electron bunch tail or accelerate a new electron bunch with properties not accessible by RF technology. Additionally, experiments on deceleration of the incoming electron bunches demonstrate the potential as dump for high energy particle beams. Such applications hold great promises for future accelerator technology and put this facility at the forefront of the current research in this field [6].

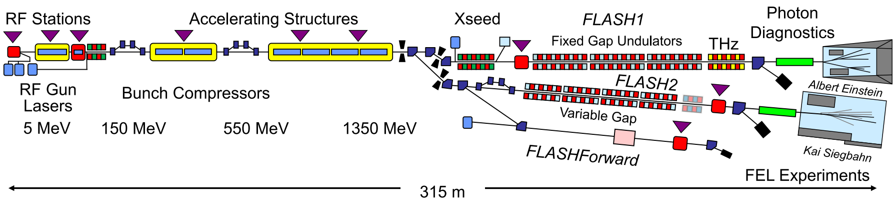

Fig. 2 Layout of the FLASH facility. Electrons are accelerated from left to right and are fed into the three electron lines FLASH1, FLASH2 and FLASHforward. Photons are produced in undulator sections in FLASH1 and FLASH2 and are delivered to photon science user experiments in the respective experimental halls, named after Albert Einstein and Kai Siegbahn, respectively.

2 Present status and scientific highlights

With its photon parameters, FLASH provides unprecedented opportunities for photon science experiments. Especially the high number of ultrashort pulses per second and the photon energy range make FLASH a facility where the pressing questions in society can be addressed in a very specific and informative way that can provide answers that are otherwise not attainable.

FLASH addresses a variety of scientific fields and its unique properties make it especially strong for femtosecond time-resolved high-resolution spectroscopy on quantum materials, in the fields of catalytic dynamics and fundamental (photo-) chemistry, as well as for charge migration and energy dissipation dynamics in biological molecules. With its pioneering role and its eased accessibility for special experimental setups, FLASH is also still strongly contributing to the development of new FEL methodologies and diagnostics.

One of the major diagnostics developments are the so-called gas monitor detectors (GMDs) that use the photon-induced ionization of (rare) gases to measure the pulse energy via the generated electron- and ion-current [7, 8].

At FLASH1, four different photon beamline ends are available for FEL experiments: beamline BL1 hosts the fixed CAMP endstation mainly for gas phase spectroscopy and imaging experiments [9], while beamline BL3 has an open port that can be overlapped with the THz beam from the undulator for THz pump-probe experiments. The beamlines PG1 and PG2 are served by a plane-grating monochromator, where PG1 hosts the fixed TRIXS endstation for resonant inelastic X-ray scattering experiments [10]. The open port of PG2 is often used for high-resolution spectroscopy experiments, mainly in the facility-operated photoemission instruments WESPE and HEXTOF [11]. In the beam line, a split-and-delay unit (SDU) is integrated [12, 13].

At FLASH2, the beam is currently delivered to three user beam lines, FL23, FL24 and FL26, while two further beamlines serve for the development of specialized diagnostics and machine studies. The beamline FL24 has an open port with flexible focussing by means of bendable mirrors. FL26 hosts the fixed installation of a reaction microscope (ReMi) provided and supported by the Max-Planck-Institute for nuclear physics in Heidelberg [14]. FL23 is currently under commissioning and features a pulse-elongation-compensating double monchromator [15]. An SDU installation upstream in the FLASH2 hall can serve both beamlines FL24 and FL23 and provides delays up to twenty picoseconds between the split FEL pulses.

Both experimental halls host a pump-probe laser that produces bursts of laser pulses synchronized to the FEL pulses. From a dedicated laser hutch, the beam is transported to laser setups next to the experiments, where further beam diagnostics, shaping and potential wavelength conversion can be performed. These setups as well as the experiments using the pump-probe laser are enclosed in laser safety tents.

Scientific applications at FLASH span a vast variety of fields and some highlight examples are given in the following.

One of the earliest experiments from FLASH demonstrated how short wavelengths can produce highly resolved diffraction images with a single light pulse that can even be so intense that the sample gets destroyed [16]. The concept was later used as one of the main arguments to develop FELs further.

In order to overlap FEL radiation with pump-probe lasers, work at FLASH established methods in order to find temporal overlap [17], measure pulse-to-pulse jitter [18] and to diagnose pulse durations and arrival times in non-invasive ways [19]. All of these methods have found their way into routine application at other FEL facilities around the world.

Using the synchronized THz und XUV sources, pioneering experiments measured details of the THz field cycles [20]. Later studies then used the THz beam to create transient states for example in magnetic films that were then selectively probed with the XUV pulses [21], while other studies created highly excited warm dense matter (WDM) states and the THz conductivity was probed [22].

In one of the experimental campaigns that produced the largest number of publications from a single beamtime, a group of scientists from the Max-Planck-Institute in Heidelberg used a setup where they split the FLASH pulse into two copies and overlapped both with a slight delay in gas targets. Correlating the transmitted spectrum with the incoming spectral distribution and the imposed delay and pulse energy, they discovered a wealth of novel effects and developed important understanding of the mechanisms in intense short wavelength light-matter interaction in the gas phase [23–31].

Another scientific field that has more recently attracted attention is to reproduce astrochemistry in the lab, since the harsh short wavelength radiation environment influences the chemical composition and abundance of molecules in space. Probing potential molecular reactions after irradiation with short wavelength pulses from FLASH thus generates insight into processes in interstellar space [32, 33].

In the field of method development, early observations of stimulated X-ray emission [34] as well as coherent sum-frequency generation [35] were reported and future applications proposed.

The high repetition rate of FLASH makes it an ideally suited tool for high-resolution electron spectroscopy from condensed matter samples. Photoemission spectroscopy with short pulses is limited in the acceptable number of generated electrons. Space charge can build up and spoil the spectral information content. Only the high repetition rate of pulses at lower pulse energy then allows to record meaningful spectra in reasonable time [36, 37]. Studies at FLASH have addressed, e.g., the field of surface catalysis [38], charge separation and transfer in organic photovoltaic compounds [39] and making movies of time-dependent, photon-triggered changes of orbitals of molecules absorbed on a surface [40].

3 Technological development / upgrade plans for the decade 2023-2033

At the current stage, FLASH is undergoing an ambitious upgrade program called FLASH2020+, within which refurbishments, improvements and changes of almost all parts of the accelerator and photon complex are performed and planned as a result of consultations with the user community [41].

In a first shutdown period, work concentrates on upgrading the accelerator part during the year 2022. Main work centers around the exchange of two of the seven accelerator modules to more modern versions that provide higher acceleration gradients. This increases the highest possible electron energy from 1.25 GeV to 1.35 GeV which in turn produces about 20% shorter photon wavelengths. Among other work, also a new injector laser system as well as a laser heater are integrated into the machine and the bunch compression sections are optimized. The installation of a variable polarization afterburner undulator at FLASH2 will provide flexibly (also circularly) polarized pulses at the third harmonic of FLASH, covering the absorption edges of elemental magnetic $3d$-materials [42].

With these new, enhanced features, FLASH is operated for users, while the next major shutdown period starting in 2024 is prepared. During that shutdown phase, the complete FLASH1 tunnel is emptied with changes all the way downstream to the experimental hall. The undulator section is completely removed to make space for the installation of external laser seeding into the two modulators necessary for echo-enabled high-harmonic generation (EEHG) [43]. With this scheme, FLASH aims for operating external seeding at photon energies up to the carbon K-edge with full coherence and high repetition rate. Anticipated photon beam parameters yield pulses of less than 10fs duration with spectral bandwidths of well below 0.1% and nearly Fourier transform limited spectral content.

The 'radiator'-undulators of APPLE-III type that generate the XUV and soft X-ray radiation will provide tunable polarization at variable gap sizes, adapting the concepts already tested at the FLASH2 afterburner undulator. The gap-tunability of the undulators at FLASH1 and FLASH2 finally removes most of the constraints to the electron energy imposed by the fixed gap undulators at FLASH1. With just a few electron energy working points, accelerator setup times are envisioned to be largely reduced and the parallel operation of FLASH1 and FLASH2 is eased.

Downstream the FLASH1 undulators, a new photon beam transport and diagnostics section will be installed, based on the state-of-the-art diagnostics at FLASH2 [44], but particularly optimized for the analysis of seeded radiation. In the experimental hall, the photon beam will be steered to the same starting point as before, such that the location of all the photon beam lines can be preserved. Small adaptations regarding few mirrors have to be made though, due to a longitudinal shift of the source point.

In the experimental halls, some further work will be devoted to optimizing the optics coatings for higher transmission at shorter wavelengths. Replacing the beam line BL3 by an improved 'FL11' beam line at shallower angles of incidence is planned. The new beam line will also be equipped with bendable KB optics for variable spot sizes and it will be optimized for transporting the higher harmonics of the FEL source as well. The integration with the THz beam line will be improved.

A further large part of the planned upgrade work concerns the pump-probe lasers. An elaborated concept has been developed that implements more standardized high-power laser sources in the laser hutches in the experimental halls of FLASH1 and FLASH2. They generate synchronized, high repetition rate 1030nm radiation at pulse durations of more than 1ps, but spectrally broadened in multi-pass cells [45, 46]. The radiation is planned to be transported to modular optical delivery stations installed close to each end-station, where the final pulse compression, shaping and wavelength tuning is performed before the beam is delivered towards the user experiment. We further aim at improved synchronization such that temporal resolutions only limited by the respective pulse durations, i.e. down to 20fs and below, become possible.

In a later stage of the project, a remodeling of the FLASH2 undulator section is envisioned to realize even more advanced schemes to generate shorter pulses in the attosecond regime. Here, concepts encompass longitudinal space-charge amplifiers, harmonic lasing ideas as well as improved multi-color operation [42].

As the next development frontier for FLASH, we see the enhancement of the number of pulses delivered per second as well as the operation at shorter wavelengths in order to provide a continuous spectral range at the facilities EuXFEL and FLASH. While more pulses can be rather easily produced by increasing the repetition rate inside the burst and retaining a similar burst duration, experimentalists do not opt for going beyond 1 MHz due to constraints in ion and electron flight times, temporal resolution of detectors and detector dead times and read-out rates. They mostly prefer an increase in burst duration or an increase in burst repetition rate, both options merging into a potential continuous wave (cw) operation of the facility. With current accelerator concepts though, such developments would demand a substantial increase of the cryogenic cooling capacities for the superconducting accelerator and they necessitate a lowering of the acceleration gradient. This would be reducing the reach towards shorter wavelengths and thus be conflicting with the facility goals.

In the current footprint of FLASH, the realization of these long-term development goals thus requires the implementation of novel acceleration concepts that promise to deliver substantially higher gradients, i.e., higher acceleration fields within shorter acceleration lengths. A promising candidate here seems to be the plasma wakefield acceleration and FLASH is optimally positioned to also pioneer this field.

4 Perspective of next scientific challenges to be addressed at the facility

The anticipated properties of FLASH after the described upgrades turn FLASH into an ideal facility to address various scientific questions. Methodologically, the fully coherent and ultra-stable pulses from an externally seeded machine will allow for novel experiments that coherently control specific light-induced dynamics, like for example chemical reactions. Furthermore, the naturally small, Fourier-limited bandwidth of the pulses, together with the flexible tunability (and the high repetition rate) will allow for highly specific and ultra-sensitive spectroscopy applications that can access minute spectroscopic changes and thus study electron dynamics in unprecedented detail. The highly stable and reproducible FEL pulses will also lower the entry barrier for new user communities since this improvement relaxes the need for complex single pulse data acquisition and analysis. With ultrashort pulses, ultrafast dynamics can be studied with unseen precision. The application possibilities are large and will be detailed below for the respective scientific fields:

In atomic, molecular and optical (AMO) sciences, some of the most important scientific questions that are targeted with free-electron lasers are the fundamental aspects of the interaction of intense short wavelength radiation with small quantum systems.

Experiments serve as a benchmark for sophisticated theoretical concepts to describe the quantum mechanics of many-body systems, following ultrafast charge migration and charge transfer processes in molecular systems, and the real-time observation of molecular reactions. To be able to benchmark theoretical concepts for intense ionizing light-matter interaction of small quantum systems, it is required to detect simultaneously electrons and ions upon ionization. Furthermore, such studies rely on full tunability of the wavelength and full control of temporal and spatial pulse shape and pulse energy.

In life science, a very intense area of research is the study of charge transfer and relaxation processes in complex molecular systems such as the primary nucleobases as the building blocks of DNA. Damage and repair mechanisms are of fundamental interest as well as wavepacket motion after controlled excitation. Ideally, the excitation is localized at a specific elemental center and probed at another center with core level spectroscopy using pulses with two different X-ray photon energies.

Condensed matter systems where quantum effects play an important role show an extremely rich variety of macroscopic physical properties as a result of a subtle interplay between electronic (charge, orbital, and spin) and lattice degrees of freedom. A possible pathway to understand the microscopic driving forces is to probe the relaxation of the relevant electronic states with ultrashort X-ray pulses after the complex systems have been excited out of equilibrium using optical light pulses. The idea is to take electronic structure maps as a function of time after a system has been driven out of equilibrium.

In general, X-ray based spectroscopic techniques allow to investigate the ultrafast response of the electronic structure in a site-selective, element-specific way and hence help to reveal the pathways from microscopic excitations to changes in macroscopic properties in these complex systems.

Light-induced ultrafast dynamics is not only of interest in the quest for a fundamental understanding of emergent phenomena in quantum materials but also because of the possibility for fast reversible switching between states with different macroscopic properties and for controlling quantum phases. The search for fingerprints of light-induced transient, metastable phases which could in turn enable one to understand and steer transitions between the different stable phases of the complex materials has attracted tremendous interest recently. For all these studies FLASH is ideally positioned with multi-color flexible pump-probe schemes and unique capabilities to monitor the evolution of electronic structure and fundamental excitations.

The short pulses at high repetition rates make FLASH, e.g., the ideal spectroscopy machine to study ultrafast dynamics in photocatalytic processes, heterogenous catalysis on surfaces and charge transfer dynamics in systems relevant for photovoltaics. It turns out that all chemical transformations, i.e., the formation and breaking of chemical bonds, are in general rare events. Each reaction partner is very rarely and for a very short time only in the critical intermediate state of a reaction. It is the availability of short, well-controlled resonant X-ray pulses at high repetition rate in combination with adequate trigger pulses that allows for the study of those rare reaction intermediates to advance our understanding how reactions proceed on their natural femtosecond time scale and how we can choose optimal reaction conditions for more energy- and resource-efficient chemistry. While for photocatalytic reactions the trigger to initiate a reaction is a femtosecond optical laser pulse, ideally resonant to the relevant electronic transition in the molecule, industrially relevant heterogeneous catalytic reactions are in general thermally activated. Here it is important to drive the key reaction pathways with the flexible pump laser pulses in the wavelength range from THz to the visible. These will be offered by FLASH so that those reactions can be triggered on an ultrashort time scale, such that a detectable fraction of reactants is found in the intermediate state at the same time.

A key topic that needs experimental exploration is the study of conical intersections: the quantum mechanically coupled crossing of different electronic states in the potential energy landscape of different atomic geometries. Most theoretical descriptions fail at conical intersections (these cannot be described by the ubiquitous Born-Oppenheimer approximation). Nevertheless, it is those crossings that determine which of the possible reaction pathways are taken by the system and a detailed understanding of the dynamics across conical intersections is thus crucial for an optimal tuning of reaction conditions to our needs. Element specific probing with near-edge X-ray absorption or resonant X-ray emission techniques after an ultrashort optical laser pulse has driven a reactant close to a conical intersection allows to directly follow the pathway of the products on the potential energy surfaces. This provides crucial input and benchmarks for the development of novel theoretical concepts and the necessary experiments can ideally be performed at FLASH.

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Data availability statement. Data sharing not applicable to this article as no datasets were generated or analysed during the current study.

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