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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.

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 all short wavelength FEL sources, FLASH is a single-pass FEL. It is based on the principle of self-amplified spontaneous emission (SASE) and 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). Electron bunches are produced 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 1990 s. 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 1990 s, 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 is 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 at FLASH1 and FLASH2 (Figs. 1 and 2).

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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

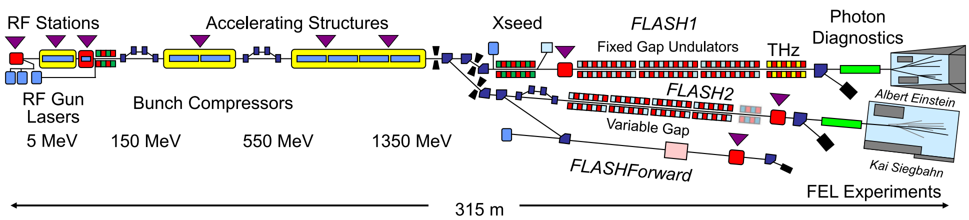

Fig. 2 Layout of the FLASH facility. Electrons are accelerated from left to right and are fed into the three electron lines FLASH1, FLASH2 and FLASH-forward. Photons are produced in undulator sections in FLASH1 and FLASH2 and are delivered to photon science user experiments in the respective experimental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

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].

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 important and impactful scientific questions can be addressed in a unique manner.

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 is 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 beamlines 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 (time-resolved resonant inelastic X-ray scattering) 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 (wide-angle electron spectrometer) and HEXTOF (high-energy X-ray time-of-flight) [11]. In the beam line, a split-and-delay unit (SDU) is integrated [12, 13].

At FLASH2, the beam is currently delivered to three beam lines for flexible experiments, 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 monochromator [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.

With regards to method development for the investigation of electronic phenomena in condensed matter samples, early observations of stimulated X-ray emission [20] as well as coherent sum-frequency generation [21] were reported and future applications proposed.

Using the synchronized THz und XUV sources, pioneering experiments measured details of the THz field cycles [22]. 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 [23], while other studies created highly excited warm dense matter (WDM) states and the THz conductivity was probed [24].

Understanding of the mechanisms in intense short wavelength light-matter interaction has been obtained in split and delay experiments. [25–30].

Recently, FLASH has increased its impact in photochemistry and astrochemistry. Experiments on thionated nucleobases in gas phase could identify the origins for UV induced triplet-state generation. The studies investigate the coupled electron-nuclear motion in the vicinity of the molecules' conical intersection [31, 32]. The core-level photoelectron spectra show charge moving from sulfur to the heterocycle and open the path to molecular electronic movies [32]. The astrochemistry community uses FLASH, to study how 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 into the origins of molecules in the interstellar space [33, 34].

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 [35, 36]. Studies at FLASH have addressed, e.g., the field of surface catalysis [37], charge separation and transfer in organic photovoltaic compounds [38] and making movies of time-dependent, photon-triggered changes of orbitals of molecules absorbed on a surface [39].

3 Technological development and 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 [40].

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 [41].

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) [42]. 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 [43], 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 [44, 45]. 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 [41].

As the next development frontier for FLASH, we see the enhancement of the number of pulses delivered per second. While more pulses can be rather easily produced by increasing the repetition rate inside the burst and retaining a similar burst duration, rates beyond 1 MHz are disadvantageous for charged particle spectroscopy because of long ion and electron flight times, restricted temporal resolution of detectors, and detector dead times and read-out rates. Many scientific communities 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 reduce the reach towards shorter wavelengths which limits many scientific fields at the facility.

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

In short, upgraded FLASH will possess the following properties, distinguishing it from the current machine:

1. an extended photon energy range up to the nitrogen K-edge in the fundamental and also deliver more photons in the high photon energy region at FLASH2
2. variable, controlled polarization in the third harmonic of FLASH2
3. an externally seeded FLASH1 up to the carbon K-edge with close to Fourier transform limited pulses of small bandwidth and high spectral and temporal stability.

The anticipated properties of FLASH after the described upgrades increase the power of FLASH to address specific scientific questions.

The extended spectral range will have an impact on element-sensitive probing of dynamics at FLASH. In the case of molecular dynamics, this will make selective probing of carbon and nitrogen much easier. So far, experiments at the nitrogen K-edge relied on the third harmonic, resulting in lower flux and also adding the complication to filter out fundamental light. Hence, experiments with dilute samples at the nitrogen K-edge become possible. This will be interesting when studying azo-photoswitch molecules. Azobenzene is available in many different variations and can be used to accomplish opti-mechanical transduction in chemical and biological samples on a single molecule scale via its optically controllable trans-cis and reverse switching [46, 47]. In addition, the spectral extension to the nitrogen edge allows for ultrafast studies on nitrogen as heteroatom in heterorganic cycles. Nucleobases are part of this class, but also interesting newly designed materials like CN compounds [48].

Variable polarization at the third harmonic of the undulator benefits spectroscopic techniques addressing the handedness in materials. In molecules, this allows addressing the molecular chirality with a combination of femtosecond time resolution and the X-ray typical site sensitivity. This leads to a novel understanding of molecular dynamics and interesting phenomena like molecular ring currents [49]. A method prone to be used is photoelectron circular dichroism, which has been established with ultrafast X-ray pulses recently [50]. In the solid state context, variable polarization addresses the magnetic subsystem of matter. The spectral range of the third harmonic undulator is ideally suited for element-selective probing of $3d$ transition metals, in particular, Fe, Ni, and Co via their *L*-edges. Circular X-ray magnetic dichroism is a well-established technique and has demonstrated its power in the ultrafast X-ray context [51].

The combination of external seeding and high repetition rate, characteristic of a superconducting accelerator, is unique in the world of FELs. In the following, we discuss a few examples where the properties of externally seeded FEL pulses present a unique advantage. As mentioned above, high repetition rate FEL radiation facilitates several applications. Electron spectroscopy on surfaces and interfaces for instance suffers kinetic energy broadening induced by space charge. Dividing the delivered photons and thus ejected electrons into many weak pulses increases the energy resolution. At SASE FEL sources, the pulses are usually filtered in a monochromator, translating to a strong shot-to-shot fluctuation, which in turn leads to wasted photons as pulses might deliver very few photons, or on the other extreme too many photons and again space charge broadening. The spectral stability of seeded pulses thus increases the signal-to-noise ratio in surface photoelectron spectroscopy. In fundamental heterogeneous catalysis studies, the new source will thus help in deciphering spectra from molecules on the surface, for instance identifying different adsorption sites or discovering small dynamical shifts after photo excitation due to geometry changes or charge transfer. The argument translates to heterogenous light-harvesting systems, such as dye-sensitized solar cells, and other molecule-functionalized surfaces [52].

In addition to the pure surface phenomena, the spectral stability arguments also hold for time-resolved spectroscopy of quantum materials with their subtle interplay between electronic (charge, orbital, and spin) and lattice degrees of freedom. The increased signal-to-noise with a seeded, high repetition rate machine can decipher the dynamic interplay of the material subsystems with higher detail.

Spectral and temporal stability provides a major advantage for time-resolved spectroscopy with observables that cannot be obtained on a single-shot base because of slow methodology or slow detector readout. Examples could include 2d-CCD cameras for capturing spectra, high-resolution mass spectroscopy, or even, usually very slow techniques that the community does not think about like chromatography or nuclear magnetic resonance. As the observable averages over many pump-probe pairs, any variation in delay and frequency would wash out information; a fact that can be avoided with a seeded FEL.

Seeding increases the spectral flux, thus benefitting methods that are 'photon-hungry' and at the same time require high spectral resolution. Typically, the SASE FLASH has a bandwidth of 0.5%. The seeded FEL FERMI in contrast provides pulses with a bandwidth of 0.05% [53]. Performing experiments with a resolution of 0.05% by inserting a monochromator into a SASE beam would diminish the spectral flux by roughly a factor of 1000. Resonant inelastic X-ray scattering (RIXS) is benefitting from the superior time-integrated spectral flux by combining the low bandwidth due to seeding with the high repetition rate of FLASH. The technique allows for the identification of quasi-particle excitations in solids [54], governing material phase transitions. In combination with the ultrashort pulses and pre-excitation with an optical laser, very sensitive probing of the materials dynamics is provided. In chemistry, RIXS has shown its potential to probe the ultrafast spin dynamics in transition metal complexes, which can be solved in high concentrations [55]. Extending the electronic sensitivity of RIXS to more dilute cases is a challenge that is alleviated by an increase in the integrated spectral flux of the FEL.

The external seeding of FLASH will create pulses with a duration close to their Fourier-transform limit. This high level of temporal coherence of FEL pulses has been exploited at FERMI for coherent control [56] and the creation of attosecond pulse trains [57]. Translating seeding-related coherence to high repetition rates opens the avenue for combining these schemes with solid and interface electron spectroscopy and dilute samples in gas and liquid phase. The increased control over pulse properties however will be a major benefit for nonlinear X-ray spectroscopy. Narrow-bandwidth pulses allow for spectrally well-resolved features in stimulated X-ray Raman spectroscopy [58, 59] and will have an application for nonlinear techniques providing increased spectral resolution when combined with spectrally broader pulses in Raman schemes [60] similar to femtosecond stimulated Raman spectroscopy in the visible spectral domain [61]. One important application of this scheme will be in the fundamental exploration of conical intersections, regions of degeneracy among potential energy surfaces, that often determine the reaction path in photochemistry. A combination of narrow and broadband coherent X-ray pulses allows for the investigation of coherences in the vicinity of conical intersections [62]. The wealth of nonlinear X-ray spectroscopic techniques with its combinations of different pulse lengths and photon energies will challenge the implementation of new FEL schemes at our facility and other FEL facilities worldwide.

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References

1. J. Rossbach, J.R. Schneider, W. Wurth, 10 years of pioneering X-ray science at the Free-Electron Laser FLASH at DESY. Phys. Rep. **808**, 1–74 (2019). https://doi.org/10.1016/j.physrep.2019.02.002
2. J. Andruszkow, B. Aune, V. Ayvazyan, N. Baboi, R. Bakker, V. Balakin, D. Barni, A. Bazhan, M. Bernard, A. Bosotti, J.C. Bourdon, W. Brefeld, R. Brinkmann, S. Buhler, J.-P. Carneiro, M. Castellano, P. Castro, L. Catani, S. Chel, Y. Cho, S. Choroba, E.R. Colby, W. Decking, P. Den Hartog, M. Desmons, M. Dohlus, D. Edwards, H.T. Edwards, B. Faatz, J. Feldhaus, M. Ferrario, M.J. Fitch, K. Flöttmann, M. Fouaidy, A. Gamp, T. Garvey, C. Gerth, M. Geitz, E. Gluskin, V. Gretchko, U. Hahn, W.H. Hartung, D. Hubert, M. Hüning, R. Ischebek, M. Jablonka, J.M. Joly, M. Juillard, T. Junquera, P. Jurkiewicz, A. Kabel, J. Kahl, H. Kaiser, T. Kamps, V.V. Katelev, J.L. Kirchgessner, M. Körfer, L. Kravchuk, G. Kreps, J. Krzywinski, T. Lokajczyk, R. Lange, B. Leblond, M. Leenen, J. Lesrel, M. Liepe, A. Liero, T. Limberg, R. Lorenz, L.H. Hua, L.F. Hai, C. Magne, M. Maslov, G. Materlik, A. Matheisen, J. Menzel, P. Michelato, W.-D. Möller, A. Mosnier, U.-C. Müller, O. Napoly, A. Novokhatski, M. Omeich, H.S. Padamsee, C. Pagani, F. Peters, B. Petersen, P. Pierini, J. Pflüger, P. Piot, B. Phung Ngoc, L. Plucinski, D. Proch, K. Rehlich, S. Reiche, D. Reschke, I. Reyzl, J. Rosenzweig, J. Rossbach, S. Roth, E.L. Saldin, W. Sandner, Z. Sanok, H. Schlarb, G. Schmidt, P. Schmüser, J.R. Schneider, E.A. Schneidmiller, H.-J. Schreiber, S. Schreiber, P. Schütt, J. Sekutowicz, L. Serafini, D. Sertore, S. Setzer, S. Simrock, B. Sonntag, B. Sparr, F. Stephan, V.A. Sytchev, S. Tazzari, F. Tazzioli, M. Tigner, M. Timm, M. Tonutti, E. Trakhtenberg, R. Treusch, D. Trines, V. Verzilov, T. Vielitz, V. Vogel, G.V. Walter, R. Wanzenberg, T. Weiland, H. Weise, J. Weisend, M. Wendt, M. Werner, M.M. White, I. Will, S. Wolff, M.V. Yurkov, K. Zapfe, P. Zhogolev, F. Zhou, First observation of self-amplified spontaneous emission in a free-electron laser at 109 nm wavelength. Phys. Rev. Lett. **85**, 3825–3829 (2000). https://doi.org/10.1103/PhysRevLett.85.3825
3. H. Wabnitz, L. Bittner, A.R.B.D. Castro, R. Döhrmann, P. Gürtler, T. Laarmann, W. Laasch, J. Schulz, A. Swiderski, K.V. Haeften, T. Möller, B. Faatz, A. Fateev, J. Feldhaus, C. Gerth, U. Hahn, E. Saldin, E. Schneidmiller, K. Sytchev, K. Tiedtke, R. Treusch, M. Yurkov, Multiple ionization of atom clusters by intense soft X-rays from a free-electron laser. Nature **420**(6915), 482–485 (2002). https://doi.org/10.1038/nature01197
4. B. Faatz, E. Plönjes, S. Ackermann, A. Agababyan, V. Asgekar, V. Ayvazyan, S. Baark, N. Baboi, V. Balandin, N.V. Bargen, Y. Bican, O. Bilani, J. Bödewadt, M. Böhnert, R. Böspflug, S. Bonfigt, H. Bolz, F. Borges, O. Borkenhagen, M. Brachmanski, M. Braune, A. Brinkmann, O. Brovko, T. Bruns, P. Castro, J. Chen, M.K. Czwalinna, H. Damker, W. Decking, M. Degenhardt, A. Delfs, T. Delfs, H. Deng, M. Dressel, H.-T. Duhme, S. Düsterer, H. Eckoldt, A. Eislage, M. Felber, J. Feldhaus, P. Gessler, M. Gibau, N. Golubeva, T. Golz, J. Gonschior, A. Grebentsov, M. Grecki, C. Grün, S. Grunewald, K. Hacker, L. Hänisch, A. Hage, T. Hans, E. Hass, A. Hauberg, O. Hensler, M. Hesse, K. Heuck, A. Hidvegi, M. Holz, K. Honkavaara, H. Höppner, A. Ignatenko, J. Jäger, U. Jastrow, R. Kammering, S. Karstensen, A. Kaukher, H. Kay, B. Keil, K. Klose, V. Kocharyan, M. Köpke, M. Körfer, W. Kook, B. Krause, O. Krebs, S. Kreis, F. Krivan, J. Kuhlmann, M. Kuhlmann, G. Kube, T. Laarmann, C. Lechner, S. Lederer, A. Leuschner, D. Liebertz, J. Liebing, A. Liedtke, L. Lilje, T. Limberg, D. Lipka, B. Liu, B. Lorbeer, K. Ludwig, H. Mahn, G. Marinkovic, C. Martens, F. Marutzky, M. Maslocv, D. Meissner, N. Mildner, V. Miltchev, S. Molnar, D. Mross, F. Müller, R. Neumann, P. Neumann, D. Nölle, F. Obier, M. Pelzer, H.-B. Peters, K. Petersen, A. Petrosyan, G. Petrosyan, L. Petrosyan, V. Petrosyan, A. Petrov, S. Pfeiffer, A. Piotrowski, Z. Pisarov, T. Plath, P. Pototzki, M.J. Prandolini, J. Prenting, G. Priebe, B. Racky, T. Ramm, K. Rehlich, R. Riedel, M. Roggli, M. Röhling, J. Rönsch-Schulenburg, J. Rossbach, V. Rybnikov, J. Schäfer, J. Schaffran, H. Schlarb, G. Schlesselmann, M. Schlösser, P. Schmid, C. Schmidt, F. Schmidt-Föhre, M. Schmitz, E. Schneidmiller, A. Schöps, M. Scholz, S. Schreiber, K. Schütt, U. Schütz, H. Schulte-Schrepping, M. Schulz, A. Shabunov, P. Smirnov, E. Sombrowski, A. Sorokin, B. Sparr, J. Spengler, M. Staack, M. Stadler, C. Stechmann, B. Steffen, N. Stojanovic, V. Sychev, E. Syresin, T. Tanikawa, F. Tavella, N. Tesch, K. Tiedtke, M. Tischer, R. Treusch, S. Tripathi, P. Vagin, P. Vetrov, S. Vilcins, M. Vogt, A.D.Z. Wagner, T. Wamsat, H. Weddig, G. Weichert, H. Weigelt, N. Wentowski, C. Wiebers, T. Wilksen, A. Willner, K. Wittenburg, T. Wohlenberg, J. Wortmann, W. Wurth, M. Yurkov, I. Zagorodnov, J. Zemella, Simultaneous operation of two soft x-ray free-electron lasers driven by one linear accelerator. New J. Phys. **18**(6), 062002 (2016). https://doi.org/10.1088/1367-2630/18/6/062002
5. R. Pan, E. Zapolnova, T. Golz, A.J. Krmpot, M.D. Rabasovic, J. Petrovic, V. Asgekar, B. Faatz, F. Tavella, A. Perucchi, S. Kovalev, B. Green, G. Geloni, T. Tanikawa, M. Yurkov, E. Schneidmiller, M. Gensch, N. Stojanovic, Photon diagnostics at the FLASH THz beamline. J. Synchrotron. Radiat. **26**(Pt 3), 700–707 (2019). https://doi.org/10.1107/s1600577519003412
6. R. D'Arcy, A. Aschikhin, S. Bohlen, G. Boyle, T. Brümmer, J. Chappell, S. Diederichs, B. Foster, M.J. Garland, L. Goldberg, P. Gonzalez, S. Karstensen, A. Knetsch, P. Kuang, V. Libov, K. Ludwig, A.M.D.L. Ossa, F. Marutzky, M. Meisel, T.J. Mehrling, P. Niknejadi, K. Pöder, P. Pourmoussavi, M. Quast, J.-H. Röckemann, L. Schaper, B. Schmidt, S. Schröder, J.-P. Schwinkendorf, B. Sheeran, G. Tauscher, S. Wesch, M. Wing, P. Winkler, M. Zeng, J. Osterhoff, FLASHForward: plasma wakefield accelerator science for high-average-power applications. Philos. Trans. R. Soc. A: Math. Phys. Eng. Sci. **377**(2151), 20180392 (2019). https://doi.org/10.1098/rsta.2018.0392
7. K. Tiedtke, J. Feldhaus, U. Hahn, U. Jastrow, T. Nunez, T. Tschentscher, S.V. Bobashev, A.A. Sorokin, J.B. Hastings, S. Möller, L. Cibik, A. Gottwald, A. Hoehl, U. Kroth, M. Krumrey, H. Schöppe, G. Ulm, M. Richter, Gas detectors for x-ray lasers. J. Appl. Phys. **103**(9), 094511 (2008). https://doi.org/10.1063/1.2913328
8. A.A. Sorokin, Y. Bican, S. Bonfigt, M. Brachmanski, M. Braune, U.F. Jastrow, A. Gottwald, H. Kaser, M. Richter, K. Tiedtke, An X-ray gas monitor for free-electron lasers. J. Synchrotron. Radiat. **26**(Pt 4), 1092–1100 (2019). https://doi.org/10.1107/s1600577519005174
9. B. Erk, J.P. Müller, C. Bomme, R. Boll, G. Brenner, H.N. Chapman, J. Correa, S. Düsterer, S. Dziarzhytski, S. Eisebitt, H. Graafsma, S. Grunewald, L. Gumprecht, R. Hartmann, G. Hauser, B. Keitel, C.V.K. Schmising, M. Kuhlmann, B. Manschwetus, L. Mercadier, E. Müller, C. Passow, E. Plönjes, D. Ramm, D. Rompotis, A. Rudenko, D. Rupp, M. Sauppe, F. Siewert, D. Schlosser, L. Strüder, A. Swiderski, S. Techert, K. Tiedtke, T. Tilp, R. Treusch, I. Schlichting, J. Ullrich, R. Moshammer, T. Möller, D. Rolles, CAMP@FLASH: an end-station for imaging, electron- and ion-spectroscopy, and pump-probe experiments at the FLASH free-electron laser. J. Synchrotron. Radiat. **25**(5), 1529–1540 (2018). https://doi.org/10.1107/s1600577518008585
10. S. Dziarzhytski, M. Biednov, B. Dicke, A. Wang, P.S.M. Miedema, R.Y. Engel, J.O. Schunck, H. Redlin, H. Weigelt, F. Siewert, C. Behrens, M. Sinha, A. Schulte, B. Grimm-Lebsanft, S.G. Chiuzbăian, W. Wurth, M. Beye, M. Rübhausen, G. Brenner, The TRIXS end-station for femtosecond time-resolved resonant inelastic x-ray scattering experiments at the soft x-ray free-electron laser FLASH. Struct. Dyn. **7**, 054301 (2020). https://doi.org/10.1063/4.0000029
11. D. Kutnyakhov, R.P. Xian, M. Dendzik, M. Heber, F. Pressacco, S.Y. Agustsson, L. Wenthaus, H. Meyer, S. Gieschen, G. Mercurio, A. Benz, K. Bühlman, S. Däster, R. Gort, D. Curcio, K. Volckaert, M. Bianchi, C. Sanders, J.A. Miwa, S. Ulstrup, A. Oelsner, C. Tusche, Y.-J. Chen, D. Vasilyev, K. Medjanik, G. Brenner, S. Dziarzhytski, H. Redlin, B. Manschwetus, S. Dong, J. Hauer, L. Rettig, F. Diekmann, K. Rossnagel, J. Demsar, H.-J. Elmers, P. Hofmann, R. Ernstorfer, G. Schönhense, Y. Acremann, W. Wurth, Time- and momentum-resolved photoemission studies using time-of-flight momentum microscopy at a free-electron laser. Rev. Sci. Instrum. **91**(1), 013109 (2020). https://doi.org/10.1063/1.5118777
12. W.F. Schlotter, F. Sorgenfrei, T. Beeck, M. Beye, S. Gieschen, H. Meyer, M. Nagasono, A. Föhlisch, W. Wurth, Longitudinal coherence measurements of an extreme-ultraviolet free-electron laser. Opt. Lett. **35**(3), 372–374 (2010). https://doi.org/10.1364/OL.35.000372
13. F. Sorgenfrei, W.F. Schlotter, T. Beeck, M. Nagasono, S. Gieschen, H. Meyer, A. Föhlisch, M. Beye, W. Wurth, The extreme ultraviolet split and femtosecond delay unit at the plane grating monochromator beamline PG2 at FLASH. Rev. Sci. Instrum. **81**, 043107 (2010). https://doi.org/10.1063/1.3374166

14. G. Schmid, K. Schnorr, S. Augustin, S. Meister, H. Lindenblatt, F. Trost, Y. Liu, M. Braune, R. Treusch, C.D. Schröter, T. Pfeifer, R. Moshammer, Reaction microscope endstation at FLASH2. J. Synchrotron. Radiat. **26**(Pt 3), 854–867 (2019). https://doi.org/10.1107/s1600577519002236
15. L. Poletto, F. Frassetto, G. Brenner, M. Kuhlmann, E. Plönjes, Double-grating monochromatic beamline with ultrafast response for FLASH2 at DESY. J. Synchrotron. Radiat. **25**(1), 131–137 (2018). https://doi.org/10.1107/s1600577517013777
16. H.N. Chapman, A. Barty, M.J. Bogan, S. Boutet, M. Frank, S.P. Hau-Riege, S. Marchesini, B.W. Woods, S. Bajt, W.H. Benner, R.A. London, E. Plönjes, M. Kuhlmann, R. Treusch, S. Düsterer, T. Tschentscher, J.R. Schneider, E. Spiller, T. Möller, C. Bostedt, M. Hoener, D.A. Shapiro, K.O. Hodgson, D.V.D. Spoel, F. Burmeister, M. Bergh, C. Caleman, G. Huldt, M.M. Seibert, F.R.N.C. Maia, R.W. Lee, A. Szöke, N. Timneanu, J. Hajdu, Femtosecond diffractive imaging with a soft-X-ray free-electron laser. Nat. Phys. **2**(12), 839–843 (2006). https://doi.org/10.1038/nphys461
17. C. Gahl, A. Azima, M. Beye, M. Deppe, K. Döbrich, U. Hasslinger, F. Hennies, A. Melnikov, M. Nagasono, A. Pietzsch, M. Wolf, W. Wurth, A. Föhlisch, A femtosecond X-ray/optical cross-correlator. Nat. Photo. **2**(3), 165–169 (2008). https://doi.org/10.1038/nphoton.2007.298
18. T. Maltezopoulos, S. Cunovic, M. Wieland, M. Beye, A. Azima, H. Redlin, M. Krikunova, R. Kalms, U. Frühling, F. Budzyn, W. Wurth, A. Föhlisch, M. Drescher, Single-shot timing measurement of extreme-ultraviolet free-electron laser pulses. New J. Phys. **10**, 033026 (2008). https://doi.org/10.1088/1367-2630/10/3/033026
19. I. Grguraš, A.R. Maier, C. Behrens, T. Mazza, T.J. Kelly, P. Radcliffe, S. Düsterer, A.K. Kazansky, N.M. Kabachnik, T. Tschentscher, J.T. Costello, M. Meyer, M.C. Hoffmann, H. Schlarb, A.L. Cavalieri, Ultrafast X-ray pulse characterization at free-electron lasers. Nat. Photo. **6**(12), 852–857 (2012). https://doi.org/10.1038/nphoton.2012.276
20. M. Beye, S. Schreck, F. Sorgenfrei, C. Trabant, N. Pontius, C. Schüßler-Langeheine, W. Wurth, A. Föhlisch, Stimulated X-ray emission for materials science. Nature **501**, 191–194 (2013). https://doi.org/10.1038/nature12449
21. H. Rottke, R.Y. Engel, D. Schick, J.O. Schunck, P.S. Miedema, M.C. Borchert, M. Kuhlmann, N. Ekanayake, S. Dziarzhytski, G. Brenner, U. Eichmann, C. von Korff Schmising, M. Beye, S. Eisebitt, Probing electron and hole colocalization by resonant four-wave mixing spectroscopy in the extreme ultraviolet. Sci. Adv. (2022). https://doi.org/10.1126/sciadv.abn5127
22. U. Frühling, M. Wieland, M. Gensch, T. Gebert, B. Schütte, M. Krikunova, R. Kalms, F. Budzyn, O. Grimm, J. Rossbach, E. Plönjes, M. Drescher, Single-shot terahertz-field-driven X-ray streak camera. Nat. Photo. **3**(9), 523–528 (2009). https://doi.org/10.1038/nphoton.2009.160
23. X. Liu, E. Jal, R. Delaunay, R. Jarrier, G.S. Chiuzbaian, G. Malinowski, T. Golz, E. Zapolnova, R. Pan, N. Stojanovic, J. Lüning, B. Vodungbo, Investigating coherent magnetization control with ultrashort THz Pulses. Appl. Sci. **12**(3), 1323 (2022). https://doi.org/10.3390/app12031323
24. Z. Chen, C.B. Curry, R. Zhang, F. Treffert, N. Stojanovic, S. Toleikis, R. Pan, M. Gauthier, E. Zapolnova, L.E. Seipp, A. Weinmann, M.Z. Mo, J.B. Kim, B.B.L. Witte, S. Bajt, S. Usenko, R. Soufli, T. Pardini, S. Hau-Riege, C. Burcklen, J. Schein, R. Redmer, Y.Y. Tsui, B.K. Ofori-Okai, S.H. Glenzer, Ultrafast multi-cycle terahertz measurements of the electrical conductivity in strongly excited solids. Nat. Commun. **12**(1), 1638 (2021). https://doi.org/10.1038/s41467-021-21756-6
25. T. Ding, M. Rebholz, L. Aufleger, M. Hartmann, K. Meyer, V. Stooß, A. Magunia, D. Wachs, P. Birk, Y. Mi, G.D. Borisova, C.D.C. Castanheira, P. Rupprecht, Z.-H. Loh, A.R. Attar, T. Gaumnitz, S. Roling, M. Butz, H. Zacharias, S. Düsterer, R. Treusch, S.M. Cavaletto, C. Ott, T. Pfeifer, Nonlinear coherence effects in transient-absorption ion spectroscopy with stochastic extreme-ultraviolet free-electron laser pulses. Phys. Rev. Lett. **123**, 103001 (2019). https://doi.org/10.1103/PhysRevLett.123.103001
26. C. Ott, L. Aufleger, T. Ding, M. Rebholz, A. Magunia, M. Hartmann, V. Stooß, D. Wachs, P. Birk, G.D. Borisova, K. Meyer, P. Rupprecht, C. da Costa Castanheira, R. Moshammer, A.R. Attar, T. Gaumnitz, Z.-H. Loh, S. Düsterer, R. Treusch, J. Ullrich, Y. Jiang, M. Meyer, P. Lambropoulos, T. Pfeifer, Strong-field extreme-ultraviolet dressing of atomic double excitation. Phys. Rev. Lett. **123**, 163201 (2019). https://doi.org/10.1103/PhysRevLett.123.163201
27. T. Ding, M. Rebholz, L. Aufleger, M. Hartmann, V. Stooß, A. Magunia, P. Birk, G.D. Borisova, D. Wachs, C.D.C. Castanheira, P. Rupprecht, Y. Mi, A.R. Attar, T. Gaumnitz, Z.-H. Loh, S. Roling, M. Butz, H. Zacharias, S. Düsterer, R. Treusch, A. Eislage, S.M. Cavaletto, C. Ott, T. Pfeifer, Measuring the frequency chirp of extreme-ultraviolet free-electron laser pulses by transient absorption spectroscopy. Nat. Commun. **12**(1), 643 (2021). https://doi.org/10.1038/s41467-020-20846-1
28. M. Rebholz, T. Ding, V. Despré, L. Aufleger, M. Hartmann, K. Meyer, V. Stooß, A. Magunia, D. Wachs, P. Birk, Y. Mi, G.D. Borisova, C.D.C. Castanheira, P. Rupprecht, G. Schmid, K. Schnorr, C.D. Schröter, R. Moshammer, Z.-H. Loh, A.R. Attar, S.R. Leone, T. Gaumnitz, H.J. Wörner, S. Roling, M. Butz, H. Zacharias, S. Düsterer, R. Treusch, G. Brenner, J. Vester, A.I. Kuleff, C. Ott, T. Pfeifer, All-XUV pump-probe transient absorption spectroscopy of the structural molecular dynamics of di-iodomethane. Phys. Rev. **11**, 031001 (2021). https://doi.org/10.1103/PhysRevX.11.031001
29. M. Rebholz, T. Ding, L. Aufleger, M. Hartmann, K. Meyer, V. Stooß, A. Magunia, D. Wachs, P. Birk, Y. Mi, G.D. Borisova, C.D.C. Castanheira, P. Rupprecht, M. Magrakvelidze, U. Thumm, S. Roling, M. Butz, H. Zacharias, S. Düsterer, R. Treusch, G. Brenner, C. Ott, T. Pfeifer, XUV-initiated dissociation dynamics of molecular oxygen (O_2). J. Phys. Chem. A **125**(47), 10138–10143 (2021). https://doi.org/10.1021/acs.jpca.1c06033
30. L. Aufleger, P. Friebel, P. Rupprecht, A. Magunia, T. Ding, M. Rebholz, M. Hartmann, C. Ott, T. Pfeifer, Line-shape broadening of an autoionizing state in helium at high XUV intensity. New J. Phys. **24**(1), 013014 (2022). https://doi.org/10.1088/1367-2630/ac3b2e
31. F. Lever, D. Mayer, D. Picconi, J. Metje, S. Alisauskas, F. Calegari, S. Düsterer, C. Ehlert, R. Feifel, M. Niebuhr, B. Manschwetus, M. Kuhlmann, T. Mazza, M.S. Robinson, R.J. Squibb, A. Trabattoni, M. Wallner, P. Saalfrank, T.J.A. Wolf, M. Gühr, Ultrafast dynamics of 2-thiouracil investigated by time-resolved Auger spectroscopy. J. Phys. B: At. Mol. Opt. Phys. **54**(1), 014002 (2020). https://doi.org/10.1088/1361-6455/abc9cb
32. D. Mayer, F. Lever, D. Picconi, J. Metje, S. Alisauskas, F. Calegari, S. Düsterer, C. Ehlert, R. Feifel, M. Niebuhr, B. Manschwetus, M. Kuhlmann, T. Mazza, M.S. Robinson, R.J. Squibb, A. Trabattoni, M. Wallner, P. Saalfrank, T.J.A. Wolf, M. Gühr, Following excited-state chemical shifts in molecular ultrafast x-ray photoelectron spectroscopy. Nat. Commun. **13**(1), 198 (2022). https://doi.org/10.1038/s41467-021-27908-y
33. J.W.L. Lee, D.S. Tikhonov, P. Chopra, S. Maclot, A.L. Steber, S. Gruet, F. Allum, R. Boll, X. Cheng, S. Düsterer, B. Erk, D. Garg, L. He, D. Heathcote, M. Johny, M.M. Kazemi, H. Köckert, J. Lahl, A.K. Lemmens, D. Loru, R. Mason, E. Müller, T. Mullins, P. Olshin, C. Passow, J. Peschel, D. Ramm, D. Rompotis, N. Schirmel, S. Trippel, J. Wiese, F. Ziaee, S. Bari, M. Burt, J. Küpper, A.M. Rijs, D. Rolles, S. Techert, P. Eng-Johnsson, M. Brouard, C. Vallance, B. Manschwetus, M. Schnell, Time-resolved relaxation and fragmentation of polycyclic aromatic hydrocarbons investigated in the ultrafast XUV-IR regime. Nat. Commun. **12**(1), 6107 (2021). https://doi.org/10.1038/s41467-021-26193-z
34. J.W.L. Lee, D.S. Tikhonov, F. Allum, R. Boll, P. Chopra, B. Erk, S. Gruet, L. He, D. Heathcote, M.M. Kazemi, J. Lahl, A.K. Lemmens, D. Loru, S. Maclot, R. Mason, E. Müller, T. Mullins, C. Passow, J. Peschel, D. Ramm, A.L. Steber, S. Bari, M. Brouard, M. Burt, J. Küpper, P. Eng-Johnsson, A.M. Rijs, D. Rolles, C. Vallance, B. Manschwetus, M. Schnell, The kinetic energy of PAH dication and trication dissociation determined by recoil-frame covariance map imaging. Phys. Chem. Chem. Phys. (2022). https://doi.org/10.1039/d2cp02252d
35. A. Pietzsch, A. Föhlisch, M. Beye, M. Deppe, F. Hennies, M. Nagasono, E. Suljoti, W. Wurth, C. Gahl, K. Döbrich, A. Melnikov, Towards time resolved core level photoelectron spectroscopy with femtosecond x-ray free-electron lasers. New J. Phys. **10**, 033004 (2008). https://doi.org/10.1088/1367-2630/10/3/033004
36. S. Hellmann, C. Sohrt, M. Beye, T. Rohwer, F. Sorgenfrei, M. Marczynski-Bühlow, M. Kalläne, H. Redlin, F. Hennies, M. Bauer, A. Föhlisch, L. Kipp, W. Wurth, K. Rossnagel, Time-resolved x-ray photoelectron spectroscopy at FLASH. New J. Phys. **14**(1), 013062 (2012). https://doi.org/10.1088/1367-2630/14/1/013062

37. M. Wagstaffe, L. Wenthaus, A. Dominguez-Castro, S. Chung, G.D.L. Semione, S. Palutke, G. Mercurio, S. Dziarzhytski, H. Redlin, N. Klemke, Y. Yang, T. Frauenheim, A. Dominguez, F. Kärtner, A. Rubio, W. Wurth, A. Stierle, H. Noei, Ultrafast real-time dynamics of CO oxidation over an oxide photocatalyst. ACS Catal. **10**(22), 13650–13658 (2020). https://doi.org/10.1021/acscatal.0c04098
38. F. Roth, M. Borgwardt, L. Wenthaus, J. Mahl, S. Palutke, G. Brenner, G. Mercurio, S. Molodtsov, W. Wurth, O. Gessner, W. Eberhardt, Direct observation of charge separation in an organic light harvesting system by femtosecond time-resolved XPS. Nat. Commun. **12**(1), 1196 (2021). https://doi.org/10.1038/s41467-021-21454-3
39. K. Baumgärtner, M. Reuner, C. Metzger, D. Kutnyakhov, M. Heber, F. Pressacco, C.-H. Min, T.R.F. Peixoto, M. Reiser, C. Kim, W. Lu, R. Shayduk, M. Izquierdo, G. Brenner, F. Roth, A. Schöll, S. Molodtsov, W. Wurth, F. Reinert, A. Madsen, D. Popova-Gorelova, M. Scholz, Ultrafast orbital tomography of a pentacene film using time-resolved momentum microscopy at a FEL. Nat. Commun. **13**(1), 2741 (2022). https://doi.org/10.1038/s41467-022-30404-6
40. M. Beye, S. Düsterer, B. Faatz, I. Hartl, C. Lechner, E. Plönjes-Palm, S.E. Schreiber, A. Schneidmiller, K. Tiedtke, R. Treusch, M. Yurkov, W. Wurth, FLASH2020+: Making FLASH Brighter, Faster and More Flexible: Conceptual Design Report (Verlag Deutsches Elektronen-Synchrotron. Hamburg (2020). https://doi.org/10.3204/PUBDB-2020-00465
41. B. Faatz, M. Braune, O. Hensler, K. Honkavaara, R. Kammering, M. Kuhlmann, E. Plönjes, J. Rönsch-Schulenburg, E. Schneidmiller, S. Schreiber, K. Tiedtke, M. Tischer, R. Treusch, M. Vogt, W. Wurth, M. Yurkov, J. Zemella, The FLASH facility: advanced options for FLASH2 and future perspectives. Appl. Sci. **7**(11), 1114 (2017). https://doi.org/10.3390/app7111114
42. P.R. Ribič, A. Abrami, L. Badano, M. Bossi, H.-H. Braun, N. Bruchon, F. Capotondi, D. Castronovo, M. Cautero, P. Cinquegrana, M. Coreno, M.E. Couprie, I. Cudin, M.B. Danailov, G.D. Ninno, A. Demidovich, S.D. Mitri, B. Diviacco, W.M. Fawley, C. Feng, M. Ferianis, E. Ferrari, L. Foglia, F. Frassetto, G. Gaio, D. Garzella, A. Ghaith, F. Giacuzzo, L. Giannessi, V. Grattoni, S. Grulja, E. Hemsing, F. Iazzourene, G. Kurdi, M. Lonza, N. Mahne, M. Malvestuto, M. Manfredda, C. Masciovecchio, P. Miotti, N.S. Mirian, I.P. Nikolov, G.M. Penco, G. Penn, L. Poletto, M. Pop, E. Prat, E. Principi, L. Raimondi, S. Reiche, E. Roussel, R. Sauro, C. Scafuri, P. Sigalotti, S. Spampinati, C. Spezzani, L. Sturari, M. Svandrlik, T. Tanikawa, M. Trovó, M. Veronese, D. Vivoda, D. Xiang, M. Zaccaria, D. Zangrando, M. Zangrando, E.M. Allaria, Coherent soft X-ray pulses from an echo-enabled harmonic generation free-electron laser. Nat. Photon. **13**(8), 555–561 (2019). https://doi.org/10.1038/s41566-019-0427-1
43. E. Plönjes, B. Faatz, M. Kuhlmann, R. Treusch, FLASH2: operation, beamlines, and photon diagnostics. AIP Conf. Proc. **1741**(1), 020008 (2016). https://doi.org/10.1063/1.4952787
44. P. Balla, A.B. Wahid, I. Sytcevich, C. Guo, A.-L. Viotti, L. Silletti, A. Cartella, S. Alisauskas, H. Tavakol, U. Grosse-Wortmann, A. Schönberg, M. Seidel, A. Trabattoni, B. Manschwetus, T. Lang, F. Calegari, A. Couairon, A. L'Huillier, C.L. Arnold, I. Hartl, C.M. Heyl, Postcompression of picosecond pulses into the few-cycle regime. Opt. Lett. **45**(9), 2572 (2020). https://doi.org/10.1364/ol.388665
45. M. Seidel, F. Pressacco, O. Akcaalan, T. Binhammer, J. Darvill, N. Ekanayake, M. Frede, U. Grosse-Wortmann, M. Heber, C.M. Heyl, D. Kutnyakhov, C. Li, C. Mohr, J. Müller, O. Puncken, H. Redlin, N. Schirmel, S. Schulz, A. Swiderski, H. Tavakol, H. Tünnermann, C. Vidoli, L. Wenthaus, N. Wind, L. Winkelmann, B. Manschwetus, I. Hartl, Ultrafast MHz-rate burst-mode pump-probe laser for the FLASH FEL facility based on nonlinear compression of PS-Level pulses from an Yb-amplifier chain. Laser Photon. Rev. **16**(3), 2100268 (2022). https://doi.org/10.1002/lpor.202100268
46. H.M.D. Bandara, S.C. Burdette, Photoisomerization in different classes of azobenzene. Chem. Soc. Rev. **41**(5), 1809–1825 (2011). https://doi.org/10.1039/c1cs15179g
47. M.-M. Russew, S. Hecht, Photoswitches: from Molecules to Materials. Adv. Mater. **22**(31), 3348–3360 (2010). https://doi.org/10.1002/adma.200904102
48. X. Wang, K. Maeda, A. Thomas, K. Takanabe, G. Xin, J.M. Carlsson, K. Domen, M. Antonietti, A metal-free polymeric photocatalyst for hydrogen production from water under visible light. Nat. Mater. **8**(1), 76–80 (2009). https://doi.org/10.1038/nmat2317
49. J.R. Rouxel, S. Mukamel, Molecular chirality and its monitoring by ultrafast x-ray pulses. Chem. Rev. **122**(22), 16802–16838 (2022). https://doi.org/10.1021/acs.chemrev.2c00115
50. M. Ilchen, P. Schmidt, N.M. Novikovskiy, G. Hartmann, P. Rupprecht, R.N. Coffee, A. Ehresmann, A. Galler, N. Hartmann, W. Helml, Z. Huang, L. Inhester, A.A. Lutman, J.P. MacArthur, T. Maxwell, M. Meyer, V. Music, H.-D. Nuhn, T. Osipov, D. Ray, T.J.A. Wolf, S. Bari, P. Walter, Z. Li, S. Moeller, A. Knie, P.V. Demekhin, Site-specific interrogation of an ionic chiral fragment during photolysis using an X-ray free-electron laser. Commun. Chem. **4**(1), 119 (2021). https://doi.org/10.1038/s42004-021-00555-6
51. I. Radu, K. Vahaplar, C. Stamm, T. Kachel, N. Pontius, H.A. Dürr, T.A. Ostler, J. Barker, R.F.L. Evans, R.W. Chantrell, A. Tsukamoto, A. Itoh, A. Kirilyuk, T. Rasing, A.V. Kimel, Transient ferromagnetic-like state mediating ultrafast reversal of antiferromagnetically coupled spins. Nature **472**(7342), 205–208 (2011). https://doi.org/10.1038/nature09901
52. N. Katsonis, M. Lubomska, M.M. Pollard, B.L. Feringa, P. Rudolf, Synthetic light-activated molecular switches and motors on surfaces. Prog. Surf. Sci. **82**(7), 407–434 (2007). https://doi.org/10.1016/j.progsurf.2007.03.011
53. E. Allaria, R. Appio, L. Badano, W.A. Barletta, S. Bassanese, S.G. Biedron, A. Borga, E. Busetto, D. Castronovo, P. Cinquegrana, S. Cleva, D. Cocco, M. Cornacchia, P. Craievich, I. Cudin, G. D'Auria, M.D. Forno, M.B. Danailov, R.D. Monte, G.D. Ninno, P. Delgiusto, A. Demidovich, S.D. Mitri, B. Diviacco, A. Fabris, R. Fabris, W. Fawley, M. Ferianis, E. Ferrari, S. Ferry, L. Froehlich, P. Furlan, G. Gaio, F. Gelmetti, L. Giannessi, M. Giannini, R. Gobessi, R. Ivanov, E. Karantzoulis, M. Lonza, A. Lutman, B. Mahieu, M. Milloch, S.V. Milton, M. Musardo, I. Nikolov, S. Noe, F. Parmigiani, G. Penco, M. Petronio, L. Pivetta, M. Predonzani, F. Rossi, L. Rumiz, A. Salom, C. Scafuri, C. Serpico, P. Sigalotti, S. Spampinati, C. Spezzani, M. Svandrlik, C. Svetina, S. Tazzari, M. Trovo, R. Umer, A. Vascotto, M. Veronese, R. Visintini, M. Zaccaria, D. Zangrando, M. Zangrando, Highly coherent and stable pulses from the FERMI seeded free-electron laser in the extreme ultraviolet. Nat. Photon. **6**(10), 699–704 (2012). https://doi.org/10.1038/nphoton.2012.233
54. L.J.P. Ament, M.V. Veenendaal, T.P. Devereaux, J.P. Hill, J.V.D. Brink, Resonant inelastic x-ray scattering studies of elementary excitations. Rev. Modern Phys. **83**(2), 705–767 (2011). https://doi.org/10.1103/revmodphys.83.705
55. P. Wernet, K. Kunnus, I. Josefsson, I. Rajkovic, W. Quevedo, M. Beye, S. Schreck, S. Grübel, M. Scholz, D. Nordlund, W. Zhang, R.W. Hartsock, W.F. Schlotter, J.J. Turner, B. Kennedy, F. Hennies, F.M.F. de Groot, K.J. Gaffney, S. Techert, M. Odelius, A. Föhlisch, Orbital-specific mapping of the ligand exchange dynamics of $fe(co)_5$ in solution. Nature **520**(7545), 78–81 (2015). https://doi.org/10.1038/nature14296
56. K.C. Prince, E. Allaria, C. Callegari, R. Cucini, G.D. Ninno, S.D. Mitri, B. Diviacco, E. Ferrari, P. Finetti, D. Gauthier, L. Giannessi, N. Mahne, G. Penco, O. Plekan, L. Raimondi, P. Rebernik, E. Roussel, C. Svetina, M. Trovo, M. Zangrando, M. Negro, P. Carpeggiani, M. Reduzzi, G. Sansone, A.N. Grum-Grzhimailo, E.V. Gryzlova, S.I. Strakhova, K. Bartschat, N. Douguet, J. Venzke, D. Iablonskyi, Y. Kumagai, T. Takanashi, K. Ueda, A. Fischer, M. Coreno, F. Stienkemeier, Y. Ovcharenko, T. Mazza, M. Meyer, Coherent control with a short-wavelength free-electron laser. Nat. Photon. **10**(3), 176–179 (2016). https://doi.org/10.1038/nphoton.2016.13
57. P.K. Maroju, C. Grazioli, M.D. Fraia, M. Moioli, D. Ertel, H. Ahmadi, O. Plekan, P. Finetti, E. Allaria, L. Giannessi, G.D. Ninno, C. Spezzani, G. Penco, S. Spampinati, A. Demidovich, M.B. Danailov, R. Borghes, G. Kourousias, C.E.S.D. Reis, F. Bille, A.A. Lutman, R.J. Squibb, R. Feifel, P. Carpeggiani, M. Reduzzi, T. Mazza, M. Meyer, S. Bengtsson, N. Ibrakovic, E.R. Simpson, J. Mauritsson, T. Csizmadia, M. Dumergue, S. Kühn, H.N. Gopalakrishna, D. You, K. Ueda, M. Labeye, J.E. Bækhøj, K.J. Schafer, E.V. Gryzlova, A.N. Grum-Grzhimailo, K.C. Prince, C. Callegari, G. Sansone, Attosecond pulse shaping using a seeded free-electron laser. Nature **578**(7795), 386–391 (2020). https://doi.org/10.1038/s41586-020-2005-6

58. T. Kroll, C. Weninger, F.D. Fuller, M.W. Guetg, A. Benediktovitch, Y. Zhang, A. Marinelli, R. Alonso-Mori, A. Aquila, M. Liang, J.E. Koglin, J. Koralek, D. Sokaras, D. Zhu, J. Kern, J. Yano, V.K. Yachandra, N. Rohringer, A. Lutman, U. Bergmann, Observation of seeded MN kβ stimulated x-ray emission using two-color x-ray free-electron laser pulses. Phys. Rev. Lett. **125**(3), 037404 (2020). https://doi.org/10.1103/physrevlett.125.037404
59. D.J. Higley, Z. Chen, M. Beye, M. Hantschmann, A.H. Reid, V. Mehta, O. Hellwig, G.L. Dakovski, A. Mitra, R.Y. Engel, T. Maxwell, Y. Ding, S. Bonetti, M. Bucher, S. Carron, T. Chase, E. Jal, R. Kukreja, T. Liu, A. Föhlisch, H.A. Dürr, W.F. Schlotter, J. Stöhr, Stimulated resonant inelastic x-ray scattering in a solid. Comm. Phys. **5**, 83 (2022). https://doi.org/10.1038/s42005-022-00857-8
60. K. Bennett, Y. Zhang, M. Kowalewski, W. Hua, S. Mukamel, Multidimensional resonant nonlinear spectroscopy with coherent broadband x-ray pulses. Phys. Scr. **T169**, 014002 (2016). https://doi.org/10.1088/0031-8949/t169/1/014002
61. P. Kukura, D.W. McCamant, R.A. Mathies, Femtosecond stimulated Raman spectroscopy. Annu. Rev. Phys. Chem. **58**(1), 461–488 (2007). https://doi.org/10.1146/annurev.physchem.58.032806.104456
62. D. Keefer, T. Schnappinger, R.D. Vivie-Riedle, S. Mukamel, Visualizing conical intersection passages via vibronic coherence maps generated by stimulated ultrafast X-ray Raman signals. Proc. Natl. Acad. Sci. **117**(39), 24069–24075 (2020). https://doi.org/10.1073/pnas.2015988117