Operation of a free-electron laser from the extreme ultraviolet to the water window

A list of authors and their affiliations appears at the end of the paper

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We report results on the performance of a free-electron laser operating at a wavelength of 13.7 nm where unprecedented peak and average powers for a coherent extreme-ultraviolet radiation source have been measured. In the saturation regime, the peak energy approached 170 μJ for individual pulses, and the average energy per pulse reached 70 μJ. The pulse duration was in the region of 10 fs, and peak powers of 10 GW were achieved. At a pulse repetition frequency of 700 pulses per second, the average extreme-ultraviolet power reached 20 mW. The output beam also contained a significant contribution from odd harmonics of approximately 0.6% and 0.03% for the 3rd (4.6 nm) and the 5th (2.75 nm) harmonics, respectively. At 2.75 nm the 5th harmonic of the radiation reaches deep into the water window, a wavelength range that is crucially important for the investigation of biological samples.

The generation of laser-like radiation in the extreme-ultraviolet (EUV) spectral range has been a grand challenge for scientists and engineers dating back to the very earliest days of the laser in the 1960s. Impressive progress was made in the 1980s, when lasing in the EUV by means of population inversion between levels in highly charged ions was observed in laser plasma experiments[1,2]. Work on these sources has undergone continuous development since then, even including a seeding option[3]. High harmonics of optical lasers were also being established at that time as sources of coherent EUV radiation[4,5]. Indeed, harmonics up to very high orders have been generated from solids recently[6]. An alternative approach, and one that is now beginning to pay rich dividends, is the production of laser-quality radiation using free electrons accelerated to relativistic speeds in linear accelerators.

Recent advances in accelerator and precision magnetic undulator technologies now make possible the construction of single-pass free-electron lasers (FELs) based on self-amplified spontaneous emission (SASE), where the light amplification process starts from shot noise in the electron beam[7–9]. These new sources provide uniquely intense, polarized, short-pulse radiation that is tunable throughout the very-UV (VUV) and X-ray wavelength range with a brilliance that exceeds both modern synchrotron radiation and laser plasma sources by many orders of magnitude in peak and average brilliance (Fig. 1).

The first demonstration of the SASE-FEL mechanism was carried out in 1997 and concerned lasing in the infrared wavelength range[10]. In September 2000, a group at Argonne National Laboratory (ANL) became the first to demonstrate saturation in a visible (390 nm) SASE FEL (ref. 11). Soon after, in September 2001, a group at DESY (Hamburg, Germany) demonstrated lasing to saturation deep into the vacuum-UV at 98 nm (refs 12 and 13). Since August 2005, the latest generation of SASE-FEL FLASH (free-electron laser in Hamburg) has been operating as a user facility[14], providing radiation at a fundamental wavelength that can be tuned from 47 nm to 13 nm. The facility is hosting many international groups that are exploiting the unique aspects of this source in projects ranging from atomic physics and materials science to biology[15–19]. Following the energy upgrade of the FLASH linear accelerator (linac) to 1 GeV in 2007, it will be possible to generate wavelengths down to 6 nm in the fundamental harmonic.

Recently, the German government approved 60% of the funding for a hard X-ray SASE-FEL user facility (the European X-Ray Free-Electron Laser (XFEL)[20,21]), the US Department of Energy (DOE) has approved the start of construction of the Linac Coherent Light Source (LCLS) at SLAC[22], and the project of the SPring-8 Compact SASE Source (SCSS) is under development in Japan[23]. LCLS and SCSS will use normal-conducting, room-temperature linacs, and the European XFEL will use superconducting accelerator technology. LCLS has entered the construction phase, with an expected completion date in 2008. It will be ready to host users in 2009. The expected commissioning phase for the SCSS is set for 2010, and the European XFEL will start operation in 2013. FLASH is the precursor for the European XFEL and so is a key platform technology on the blueprint for large-scale, X-ray FELs.

Brilliance, coherence and timing down to the femtosecond regime are the three properties that provide the potential for new science to be explored for EUV and X-ray FELs. Because SASE-FELs can produce pulses of unprecedented peak intensity, they open up the potential for EUV and X-ray nonlinear optics and spectroscopy. The primary interaction is with inner shell electrons, so EUV laser matter interactions will form a rich field of new science and applications in the coming decade. The first experimental results demonstrating the ability of future XFELs to generate pulses of a few tens of femtoseconds have been achieved recently at FLASH[12–14].

The timing of this report is appropriate for several reasons. A crucial milestone on the X-ray FEL roadmap was reached recently when FLASH produced saturated laser-like EUV radiation pulses at 13.7 nm for the first time. This 13 nm region is important because of its relevance to EUV lithography. At saturation, FLASH delivers ultrashort pulses with durations as low as 10 fs, and with peak and average powers of up to 10 GW and 20 mW, respectively (record values for EUV lasers). FLASH also produces bright emission at the third harmonic (4.6 nm) and the fifth harmonic (2.75 nm) of the fundamental mode. The latter wavelength is shorter than any produced so far by plasma-based X-ray lasers, and it lies well within the so-called water window where biological systems can be imaged and analysed *in vitro* (and potentially *in vivo*). In addition, the pulse durations of the harmonics decrease with harmonic number, so their durations lie in the single-digit femtosecond range, opening up the possibility of studying deep inner-shell atomic and molecular dynamics on a subfemtosecond timescale.

RESULTS

PRODUCTION OF ELECTRON BUNCHES

FLASH is a SASE FEL that produces EUV radiation during a single pass of an electron beam through a long periodic magnetic undulator[7–9]. The driving mechanism of a FEL is the radiative instability of the electron beam due to the collective interaction of electrons with the electromagnetic field in the undulator[24]. The amplification process in SASE FELs starts from the shot noise in the electron beam. When the electron beam enters the undulator, the beam modulation at wavelengths close to the resonance wavelength,

$$\lambda = \lambda_w(1+K^2)/(2\gamma^2) \qquad (1)$$

initiates the process of radiation emission (here λ_w is the undulator period, $K = eB_w\lambda_w/2\pi m_e c$ is the undulator parameter, B_w is the r.m.s. value of the undulator field, γ is the relativistic factor, c is the velocity of light and m_e and e are the mass and charge of the electron, respectively). The interaction between electrons oscillating in the undulator and the radiation that they produce, leads to a periodic longitudinal density modulation (microbunching) with a period equal to the resonance wavelength. The radiation emitted by the microbunches is in phase and adds coherently, leading to an increase in the photon intensity that further enhances the microbunching. The amplification process develops exponentially with the undulator length, and an intensity gain in excess of 10^7 is obtained in the saturation regime. At this level, the shot noise of the electron beam is amplified up to the point at which complete microbunching is achieved and almost all electrons radiate in phase, producing powerful, coherent radiation.

A qualitative estimation of the FEL operating parameter space can be obtained in terms of the FEL parameter ρ (ref. 25).

$$\rho = \left[\frac{I}{I_A}\frac{A_{JJ}^2 K^2 \lambda_w^2}{32\pi^2\gamma^2\sigma_\perp^2}\right]^{1/3}. \qquad (2)$$

Here I is the beam current, $I_A = 17$ kA is the Alfven current, $\sigma_\perp$ is the r.m.s transverse size of the electron bunch, and the coupling factor is $A_{JJ} = 1$ for a helical undulator and $A_{JJ} = [J_0(Q) - J_1(Q)]$ for a planar undulator, where $Q = K^2/[2(1+K^2)]$ and J_0 and J_1 are the Bessel functions of the first kind. Estimates for the main FEL parameters are as follows: the field gain length, $L_g \approx \lambda_w/(4\pi\rho)$, the FEL efficiency in the saturation regime is approximately equal to ρ, the spectral bandwidth is approximately 2ρ, and the coherence time is $\tau_c \approx L_g\lambda/(\lambda_w c)$.

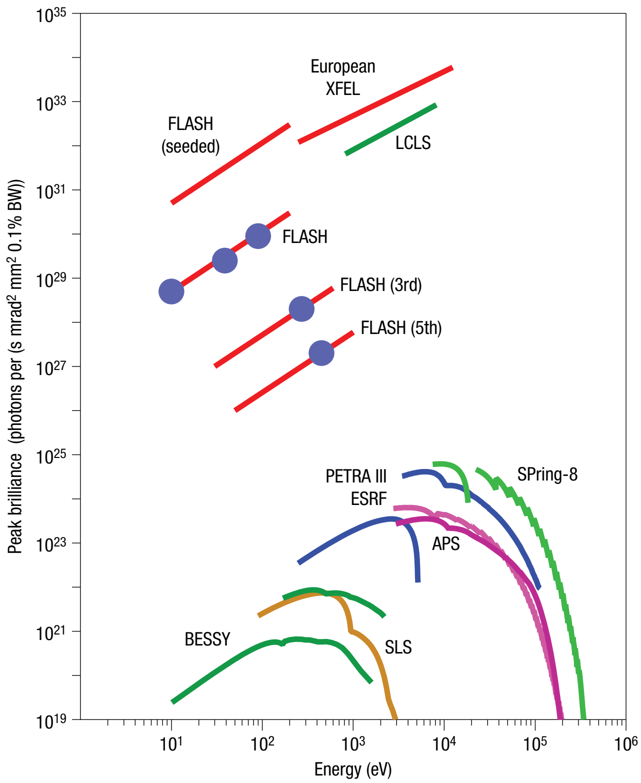

Figure 1 Peak brilliance of X-ray FELs in comparison with third-generation synchrotron-radiation light sources. Blue spots show experimental performance of the FLASH FEL at DESY at the fundamental, 3rd and 5th harmonics.

The FLASH facility has already been described in detail elsewhere[14]. A comprehensive description of specific systems, with relevant references, is presented in the Supplementary Information, (Sections 1–3). Figure 2a shows the schematic layout of the FLASH facility. The electron beam is produced in a radio-frequency gun and brought up to an energy of 700 MeV by five accelerating modules ACC1 to ACC5 (ref. 14). At energies of 130 and 380 MeV, the electron bunches are compressed in the bunch compressors BC1 and BC2. The undulator is a fixed 12-mm gap permanent magnet device with a period length of 2.73 cm and a peak magnetic field of 0.47 T. The undulator system is subdivided into six segments, each 4.5 m long.

The electron beam formation system is based on the use of nonlinear longitudinal compression. When the bunch is accelerated off-crest in the accelerating module, the longitudinal phase space acquires a radio-frequency-induced curvature. Downstream of each bunch compressor, this distortion results in a non-gaussian distribution within the bunch and in a local charge concentration. It is the leading edge of the bunch, with its high peak current, that is capable of driving the high-intensity lasing process (Fig. 2). With proper optimization of the bunch compression system, it is possible to obtain a low transverse emittance for the high-current spike, which is absolutely crucial for the production of high-quality FEL beams. In this regard, it should be noted that collective effects play a significant role in the bunch compression process for short pulses. In the high-current part of the bunch, with r.m.s length σ_z and peak current I, coherent synchrotron radiation (CSR) and longitudinal space charge (LSC) effects scale as $I/\sigma_z^{1/3}$ and I/σ_z, respectively. For instance, the LSC-induced

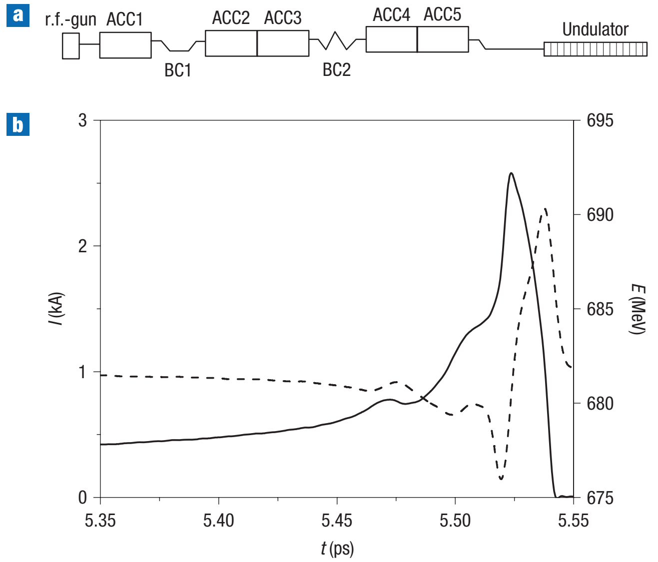

Figure 2 Production of electron bunches at FLASH. a, Schematic of the FLASH facility. Abbreviations ACC and BC stand for accelerating module and bunch compressor, respectively. The electron beam originates from the laser-driven, radio-frequency (r.f.) gun, and is accelerated and compressed to a high value of peak current in the magnetic compressors. **b**, Current, *I*, (solid line) and mean energy *E* (dashed line) along the electron bunch at the undulator entrance (simulation). The bunch head is located at the right-hand side of the figure.

energy chirp along the high-current spike of the bunch grows when the bunch travels from the bunch compressor to the undulator entrance[26]:

$$\frac{\mathrm{d}(\Delta\gamma)}{\mathrm{d}z} \approx 2.4\frac{I}{I_\mathrm{A}}\frac{\ln(\gamma\sigma_z/\sigma_\perp)}{\sigma_z\gamma^2}. \quad (3)$$

One can see the power of this effect in Fig. 2, where an increase in the peak current and a narrowing of the width of the spike leads to an increase in the induced energy chirp due to space charge effects. For FLASH, operating at a wavelength of 13 nm, the value of the FEL parameter is $\rho = (2.5 - 3) \times 10^{-3}$. The lasing portion of the electron bunch possesses a strong energy chirp (see Fig. 2b), which results in an additional broadening of the FEL spectral distribution. Another consequence of the energy chirp is suppression of the FEL gain. The effect of the energy chirp on the gain is given by the parameter $(\rho\gamma/\tau_c)^{-1}\,\mathrm{d}\gamma/\mathrm{d}t$, and it starts to play a significant role when the relative energy change acquired within one coherence length $c\tau_c$, becomes comparable with the FEL parameter ρ (refs 27 and 28). This effect is strong in the case under study here, and results in a significant correction to the FEL gain. Finally, the effect of the energy chirp leads to further shortening of the lasing part of the electron bunch (and hence the FEL pulse length) in the regime of exponential growth.

SATURATED OUTPUT AND GAIN LENGTH

So far, FLASH has demonstrated its ability to generate powerful, coherent, continuously tunable radiation in the wavelength range from 47 to 13 nm at the fundamental harmonic, and down to 2.75 nm at the 5th harmonic. In this paper we present a thorough measurement and characterization of the properties of the FEL radiation at a wavelength of 13.7 nm. The average energy in the radiation pulse demonstrated at this wavelength was 70 μJ, although the characterization reported here was performed at an average energy of 40 μJ. Figure 3 shows the spatial profile of the FEL radiation detected on a Ce:YAG screen located 23.5 m downstream of the undulator exit. The full width at half maximum (FWHM) spot size is 2.1 mm, which corresponds to an angular divergence of 90 ± 10 μrad (FWHM). The radiation mode in the far-field is nearly axisymmetric. With the additional knowledge of the EUV spot size at the undulator exit (~160 μm FWHM), we find that the product of the spot size of the radiation at the undulator exit and the angular divergence results in a value of about the radiation wavelength. This means that the phase volume of the radiation is close to the diffraction limit, and that the radiation has a high degree of transverse coherence.

We present in Fig. 4a the average EUV pulse energy versus undulator length, where the exponential growth from spontaneous radiation to saturated laser output can be seen. Each point in this figure was obtained by averaging over 300 pulses (see Supplementary Information, Section 5, for more details). The radiation energy was measured with a microchannel-plate-(MCP-) based detector[29], which was operated with a 5-mm-diameter aperture and was located 18.5 m from the undulator. The interaction length in the undulator (and hence amplification) was changed by means of a transverse kick of the electron-beam trajectory between the undulator modules, which is strong enough to stop the FEL amplification process downstream of the orbit kick. With the FEL interaction suppressed along the whole length of the undulator chain, the residual spontaneous emission from the full undulator length and also the full electron bunch was measured. The FEL interaction was switched on gradually along the undulator, and the energy in the radiation pulse grew steadily. Independent measurements of the radiation energy at saturation were made with the aid of the gas monitor detector[30]. Analysis of the exponential part of the gain curve presented in Fig. 4a yields a field gain length of $L_\mathrm{g} = 2.5 \pm 0.3$ m. This measurement gives us an estimate for the coherence time $\tau_\mathrm{c} \approx 4.2 \pm 0.5$ fs.

EUV PULSE DURATION

The energy of each EUV pulse fluctuates from shot to shot, because the lasing process starts up from shot noise. When the FEL amplification process takes place, fluctuations in the EUV field grow with undulator length and reach a maximum value at the end of the regime of exponential growth, just before saturation. When the amplification process finally reaches saturation, the fluctuations drop sharply. The measured probability distributions of the radiation energy for these two regimes are shown in Fig. 4b and c (see Supplementary Information, Section 5, for more details). Data for the exponential and the saturation regime were taken at the average energy in the radiation pulse of 1 and 40 μJ, respectively. For a SASE-FEL operating in the regime of exponential growth, it is well known that the radiation exhibits the properties of completely chaotic polarized light[31]. One consequence is that the probability distribution of the energy E, in the radiation pulse fits to a gamma distribution,

$$p(E) = \frac{M^M}{\Gamma(M)}\left(\frac{E}{\langle E\rangle}\right)^{M-1}\frac{1}{\langle E\rangle}\exp\left(-M\frac{E}{\langle E\rangle}\right) \quad (4)$$

where $\Gamma(M)$ is the gamma function, $M = 1/\sigma_E^2$, and $\sigma_E^2 = \langle(E - \langle E\rangle)^2\rangle/\langle E\rangle^2$. The parameter M can be interpreted as the average number of 'degrees of freedom' or 'modes' in the radiation pulse. Thus, from the experimental result for $M = 1.9$, and an estimate for the coherence time $\tau_c \approx 4$ fs, we come to an

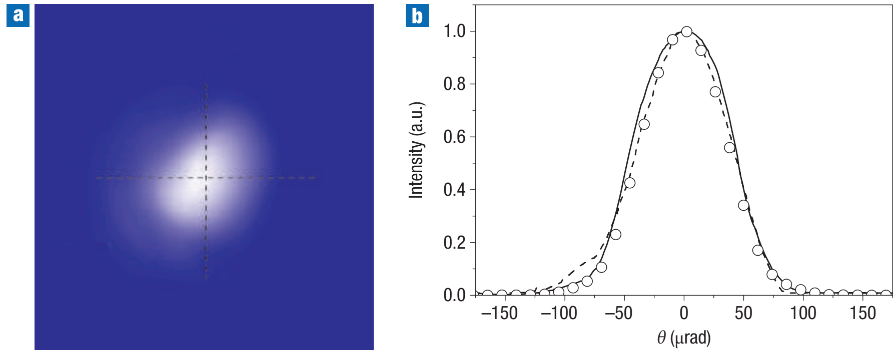

Figure 3 Spatial profile of the FEL radiation. a, Photon beam image on a Ce:YAG crystal averaged over many shots. The Ce:YAG screen is located 23.5 m downstream of the undulator exit. The FWHM spot size is 2.1 mm. **b**, Vertical (solid line) and horizontal (dashed line) slices of the image. Circles represent simulation results with the code FAST (ref. 38). The average energy in the radiation pulse is 40 μJ. The radiation wavelength is 13.7 nm.

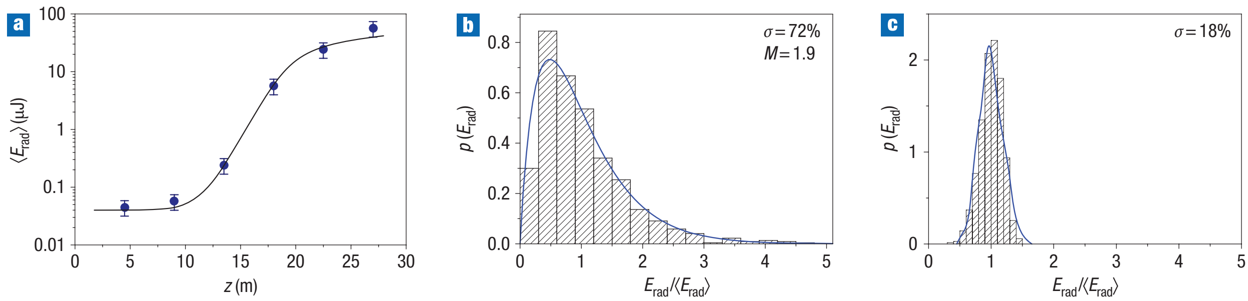

Figure 4 Energy in the radiation pulse and its fluctuations. a, Average energy in the radiation pulse vs. undulator length, showing exponential growth and saturation. Error bars correspond to the uncertainty in the calibration of the MCP detector of $\pm 15\%$. **b**,**c**, Probability distributions for the energy in the radiation pulses for the end of the regime of exponential growth (**b**) and for the saturation regime (**c**) respectively. Radiation wavelength is 13.7 nm. The solid line in **b** represents a gamma distribution, $p(E)$, given in (4) with the parameter $M = 1.9$. The solid line in **c** represents simulations with the code FAST (ref. 38).

estimate for the radiation pulse length at the end of the regime of exponential growth of about 8 ± 1 fs.

SPECTRAL DISTRIBUTION OF THE FLASH OUTPUT

Results of spectral measurements are presented in Fig. 5. During these measurements, FLASH operated in the saturation regime with an average energy in the radiation equal to 40 μJ. Single-shot spectra were obtained with the plane grating monochromator, operating in spectrographic mode, with a resolving power $\lambda/\Delta\lambda$ of 1,500 (ref. 32). The bold curve in Fig. 5a represents the average value of 300 pulses—single shots are also shown as light curves. We observe that the single-shot spectra are dominated by a single feature, albeit accompanied in some cases by some small modulations. Because the spectrum is simply a Fourier transform of the temporal structure, we conclude that the temporal profile of the radiation pulses also consists mostly of a single spike. This qualitative observation is in good agreement with the measured number of modes $M = 1.9$ (Fig. 4b), which tells us that the radiation has nearly complete longitudinal coherence. This conclusion is also supported by significant suppression of the measured fluctuations of the radiation energy when spectrally filtered by a narrow-band monochromator (see Supplementary Information, Section 6, for more details).

Another observation is the rather large width of the averaged spectrum, about 1% of the EUV wavelength. Our analysis shows that half of this value comes from the amplification bandwidth, and the additional broadening, is due to a strong energy chirp along the lasing spike (Fig. 2).

HARMONICS OF THE 13.7-NM FUNDAMENTAL MODE

Radiation from a SASE FEL operating at saturation also contains relatively strong contributions from higher-frequency harmonics[33–35]. Figure 5b shows the average spectrum of the 3rd harmonic. Comparison of the spectra of the fundamental and the 3rd harmonic shows that the relative bandwidth, $\Delta\lambda/\lambda_h$, remains nearly constant, a result that is in good agreement with theoretical predictions for the properties of higher harmonics generated in the SASE-FEL operating in the saturation regime[36]. Note that in the case of ordinary undulator radiation, the relative spectral width, $\Delta\lambda/\lambda_h$ scales inversely with the harmonic number h. The spectrum of the 5th harmonic is shown in Fig. 5c. The spectrum was obtained by averaging more than 1,000 pulses as the grating and mirror reflectivities were very small at this wavelength.

For an average EUV pulse energy of 40 μJ at the fundamental wavelength, we measured 0.25 ± 0.1 μJ for the 3rd (4.6 nm) and 10 ± 4 nJ for the 5th (2.75 nm) harmonic. These values correspond to a relative contribution to the total EUV output energy of $0.6 \pm 0.2\%$ and $0.03 \pm 0.01\%$ for the 3rd and the 5th harmonics, which is in agreement with prediction[36]. The physical mechanism underlying the generation of higher harmonics is a

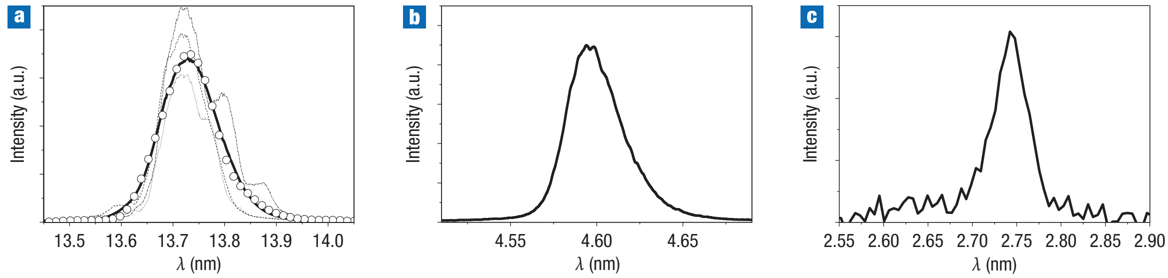

Figure 5 EUV spectra of the fundamental (a), 3rd harmonic (b) and 5th harmonic (c) contributions to the FEL output. Bold lines show averaged spectra (mean value of 300 shots), and thin lines show single-shot spectra. Circles in **a** represent averaged spectra simulated with the code FAST[38]. The average energy in the FEL pulse is 40 μJ.

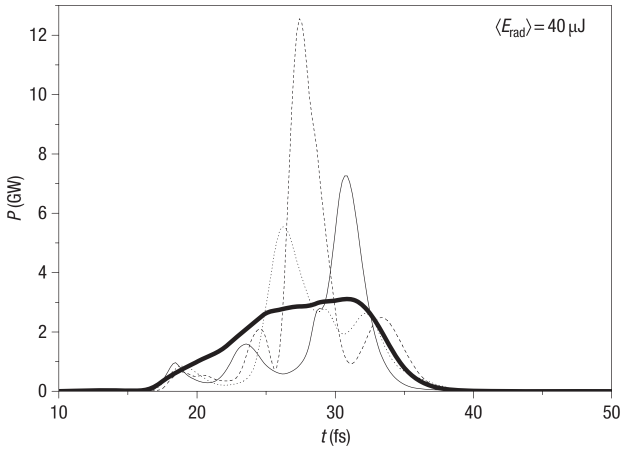

Figure 6 Temporal structure of the radiation pulse for an average energy of 40 μJ, predicted by the code FAST using experimentally determined radiation properties. The radiation wavelength is 13.7 nm. The bold line shows the envelope and the thin lines are single shots.

nonlinear transformation, which results in a shortening of the FEL pulse width. With a 10-fs pulse duration for the fundamental harmonic, we estimate peak powers of 40 MW and 2 MW for the 3rd and 5th harmonics, respectively. The corresponding average powers were approximately 120 μW and 6 μW. Hence, the available intensities are sufficient for performing experiments in the so-called 'water window', a wavelength range that is crucially important for the investigation of biological samples.

Theoretical predictions tell us that the contribution of the 2nd harmonic to the total radiation power depends strongly on the ratio of the FEL gain length to the Rayleigh length of the radiation[37]. For a ratio of unity, the 2nd harmonic intensity comes out to be a fraction of a per cent, and decreases rapidly as this ratio increases. The estimated ratio for the parameter range of FLASH operating at the wavelength of 13.7 nm is about ten. Measurements of the even harmonics have shown that they are significantly suppressed with respect to the odd harmonics. For instance, the intensity of the 2nd harmonic is more than an order of magnitude less than that of the 3rd harmonic.

With the measured properties of the radiation (gain curve, pulse energy, spectra, statistical properties and spatial properties of the radiation), we can now determine the range of electron beam parameters related to the lasing fraction of the electron bunch. The electron bunch shape and energy chirp is close to that shown in Fig. 2, with a peak current of 2–2.5 kA. The FWHM length of the high-current spike is approximately 30 fs, and the normalized emittance is 1–1.5 mm-mrad. The excellent agreement between experiment and simulation with the code FAST (ref. 38) (see Figs 3–5) allows us to specify those parameters of the radiation that cannot be measured directly. For instance, in Fig. 6 we present the temporal profile of the radiation pulse deduced from the same simulation data. We see that the pulse duration is indeed about 10 fs, in good agreement with the experimentally deduced result. The mean value of the peak power in the radiation pulse is more than 3 GW, and for some individual high-energy pulse, the peak power approaches 10 GW.

DISCUSSION

An important lesson from FLASH is that GW-level, laser-like EUV radiation pulses on a 10-fs scale can be produced with a reliable single-pass SASE-FEL scheme. FLASH has produced unprecedented powers for EUV radiation at a fundamental wavelength of 13.7 nm, and harmonics with wavelengths as low as 2.75 nm (that is, in the range of a soft X-ray FEL). The experimental measurements show that FLASH is now operating at its ultimate performance level, with a peak brilliance of $(6 \pm 3) \times 10^{29}$, $(2 \pm 1) \times 10^{28}$ and $(2 \pm 1) \times 10^{27}$ photons per (s mrad^2 mm^2 0.1% bandwidth) at 13.7, 4.6 and 2.75 nm, respectively (Fig. 1). At the 5th harmonic wavelength, FLASH is already approaching the wavelength range of the European XFEL and LCLS, albeit with lower brilliance. However, it is still higher than the peak brilliance of the third-generation SR sources by quite a few orders of magnitude.

METHODS

EUV DIAGNOSTICS

The FEL-beam diagnostics have already been described in detail elsewhere (see ref. 39 and Supplementary Information, Section 4) and so only a brief summary is given here. The FEL is operated at 5 Hz in either single-bunch or in multibunch mode, at present. In the latter mode, the FEL typically produces up to 140 radiation pulses (five times per second). The FEL pulse energies were measured using MCPs (ref. 29) and gas monitor detectors[30]. The EUV spectra of the FEL fundamental mode and its harmonics were measured using three different grazing incidence spectrometer systems. The first spectrometer is described in detail in ref. 40. Briefly, the system contains a spherical variable line spacing grating that provides a flat field in the focal plane for easy matching to a flat detector array combined with a spherical mirror in a Kirkpatrick–Baez configuration, resulting in a stigmatic spectrometer. The spectral resolution,

$\lambda/\Delta\lambda$, is better than 1,500 across the 3–40 nm operating wavelength range. The detection system consists of a phosphor faceplate for EUV to optical image down-conversion, with a readout by a lens coupled intensified CCD camera. The second spectrometer is a plane grating monochromator equipped with a 200 lines per millimetre grating for the photon wavelength range of 6–60 nm, and a high-resolution 1,200 lines per millimetre grating for the wavelength range 2–12 nm (ref. 32). These two systems were used to measure spectra of the fundamental and the third harmonic FEL pulses and, with their gated readout, they could select a single pulse from a FEL pulse train. The third monochromator was a flat field system, equipped with a Harada grating[41], but with a highly sensitive back-illuminated CCD camera (Andor Technology) containing 2,048 × 512 pixels, each with a footprint of 13 × 13 μm^2. The system was operated without an entrance slit, which resulted in a spectral resolution of approximately 100. A 1.5-μm thick aluminium foil placed over the entrance aperture provided a transmission window of 1 nm FWHM, centred at about 2.8 nm, when combined with the reflectivity of the beamline mirrors. This system was used to measure the spectrum of the 5th harmonic.

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Correspondence and requests for materials should be addressed to M.V.Y.
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W. ACKERMANN[1], G. ASOVA[2], V. AYVAZYAN[3], A. AZIMA[3], N. BABOI[3], J. BÄHR[2], V. BALANDIN[3], B. BEUTNER[4], A. BRANDT[3], A. BOLZMANN[5], R. BRINKMANN[3], O. I. BROVKO[6], M. CASTELLANO[7], P. CASTRO[3], L. CATANI[8], E. CHIADRONI[8], S. CHOROBA[3], A. CIANCHI[8], J. T. COSTELLO[9], D. CUBAYNES[10], J. DARDIS[9], W. DECKING[3], H. DELSIM-HASHEMI[4], A. DELSERIEYS[11], G. DI PIRRO[7], M. DOHLUS[3], S. DÜSTERER[3], A. ECKHARDT[4], H. T. EDWARDS[12], B. FAATZ[3], J. FELDHAUS[3], K. FLÖTTMANN[3], J. FRISCH[13], L. FRÖHLICH[4], T. GARVEY[14], U. GENSCH[2], CH. GERTH[3], M. GÖRLER[3], N. GOLUBEVA[3], H.-J. GRABOSCH[2], M. GRECKI[15], O. GRIMM[3], K. HACKER[3,4], U. HAHN[3], J. H. HAN[3], K. HONKAVAARA[4], T. HOTT[3], M. HÜNING[3], Y. IVANISENKO[16], E. JAESCHKE[17], W. JALMUZNA[18], T. JEZYNSKI[15], R. KAMMERING[3], V. KATALEV[3], K. KAVANAGH[9], E. T. KENNEDY[9], S. KHODYACHYKH[2], K. KLOSE[2], V. KOCHARYAN[3], M. KÖRFER[3], M. KOLLEWE[3], W. KOPREK[18], S. KOREPANOV[2], D. KOSTIN[3], M. KRASSILNIKOV[2], G. KUBE[3], M. KUHLMANN[3], C. L. S. LEWIS[11], L. LILJE[3], T. LIMBERG[3], D. LIPKA[3], F. LÖHL[4], H. LUNA[9], M. LUONG[19], M. MARTINS[4], M. MEYER[10], P. MICHELATO[20], V. MILTCHEV[4], W. D. MÖLLER[3], L. MONACO[20], W. F. O. MÜLLER[1], O. NAPIERALSKI[15], O. NAPOLY[19], P. NICOLOSI[21], D. NÖLLE[3], T. NUÑEZ[3], A. OPPELT[2], C. PAGANI[20], R. PAPARELLA[19], N. PCHALEK[3,4], J. PEDREGOSA-GUTIERREZ[9], B. PETERSEN[3], B. PETROSYAN[2], G. PETROSYAN[3], L. PETROSYAN[3], J. PFLÜGER[3], E. PLÖNJES[3], L. POLETTO[2], K. POZNIAK[18], E. PRAT[3,4], D. PROCH[3], P. PUCYK[18], P. RADCLIFFE[3], H. REDLIN[3], K. REHLICH[3], M. RICHTER[22], M. ROEHRS[3,4], J. ROENSCH[4], R. ROMANIUK[18], M. ROSS[13], J. ROSSBACH[4], V. RYBNIKOV[3], M. SACHWITZ[2], E. L. SALDIN[3], W. SANDNER[23], H. SCHLARB[3], B. SCHMIDT[3], M. SCHMITZ[3], P. SCHMÜSER[4], J. R. SCHNEIDER[3], E. A. SCHNEIDMILLER[3], S. SCHNEPP[1], S. SCHREIBER[3], M. SEIDEL[3,24], D. SERTORE[20], A. V. SHABUNOV[6], C. SIMON[19], S. SIMROCK[3], E. SOMBROWSKI[3], A. A. SOROKIN[25,22], P. SPANKNEBEL[26], R. SPESYVTSEV[16], L. STAYKOV[2], B. STEFFEN[3], F. STEPHAN[2], F. STULLE[3], H. THOM[3], K. TIEDTKE[3], M. TISCHER[3], S. TOLEIKIS[3], R. TREUSCH[3], D. TRINES[3], I. TSAKOV[27], E. VOGEL[3], T. WEILAND[1], H. WEISE[3], M. WELLHÖFER[4], M. WENDT[3,12], I. WILL[23], A. WINTER[3], K. WITTENBURG[3], W. WURTH[4], P. YEATES[9], M. V. YURKOV[3]*, I. ZAGORODNOV[3] AND K. ZAPFE[3]

Affiliations for authors: [1]Technische Universität Darmstadt, FB 18, Institut TEMF, Schlossgartenstr. 8, 64289 Darmstadt, Germany; [2]Deutsches Elektronen-Synchrotron DESY, Platanenallee 6, 15738 Zeuthen, Germany; [3]Deutsches Elektronen-Synchrotron DESY, Notkestrasse 85, 22603 Hamburg, Germany; [4]Universität Hamburg, Institut für Experimentalphysik, Luruper Chaussee 149, 22761 Hamburg, Germany; [5]Bayerische Julius-Maximilians Universität, Institut für Theor. Physik und Astrophysik, Am Hubland, 97074 Wurzburg, Germany; [6]Joint Institute for Nuclear Research, 141980 Dubna, Moscow Region, Russia; [7]INFN-LNF, via E. Fermi 40, 00044 Frascati, Italy; [8]INFN-Roma Tor Vergata, Via della Ricerca Scientifica, 1, 00133 Rome, Italy; [9]National Center for Plasma Science and Technology and School for Physical Sciences, Dublin City University, Dublin, Ireland; [10]LIXAM/CNRS, Centre Universitaire Paris-Sud, Batiment 350, F-91405 Orsay Cedex, France; [11]International Research Centre for Experimental Physics, Queen's University, Belfast, BT7 1NN, UK; [12]Fermi National Accelerator Laboratory, MS 306, P.O.Box 500, Batavia, Illinois 60510, USA; [13]Stanford Linear Accelerator Center, 2575 Sand Hill Road, Menlo Park, California 94025, USA; [14]Laboratoire de L'Accelerateur Lineaire, IN2P3-CNRS, Universite de Paris-Sud, B.P. 34, 91898 Orsay, France; [15]Technical University of Lodz, Al. Politechniki 11, Lodz, Poland; [16]Kharkiv National University, 4 Svobody sq, 61077, Kharkiv, Ukraine; [17]BESSY GmbH, Albert-Einstein-Str.15, 12489 Berlin, Germany; [18]Institute of Electronic Systems, Warsaw University of Technology, Nowowiejska 15/19, Warsaw, Poland; [19]CEA Saclay, 91191 Gif-sur-Yvette, France; [20]INFN Milano-LASA, via Fratelli Cervi 201,I 20090 Segrate (MI), Italy; [21]CNR-INFM, Department of Information Engineering, University of Padova, via Gradenigo 6/B, 35131, Padova, Italy; [22]Physikalisch-Technische Bundesanstalt, PTB, Abbestrasse 2-12, D-10587 Berlin, Germany; [23]Max-Born-Institute, Max-Born-Str. 2a, 12489 Berlin, Germany; [24]Paul Scherrer Institute, 5232 Villigen PSI, Switzerland; [25]Ioffe Physico-Technical Institute RAS, Polytekhnicheskaya 26, 194021 St. Petersburg, Russia; [26]Humboldt Universität, Unter den Linden 6, 10099 Berlin, Germany; [27]Institute for Nuclear Researches and Nuclear Energy, Tzarigradsko Shaussee Boulevard 72, 1784 Sofia, Bulgaria; *email: mikhail.yurkov@desy.de