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WP2: FEL Science Requirements and Facility Design

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Science Requirements and Performance Specification for the CompactLight X-Ray Free-Electron Laser

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1 Introduction

CompactLight [1] is a consortium funded by the European Union through the Horizon 2020 Research and Innovation Programme under Grant Agreement No. 777431. The 24 partner institutes are working collaboratively towards the conceptual design of a next-generation x-ray free-electron laser (FEL). CompactLight intends to design an x-ray FEL facility beyond today's state of the art, using the latest concepts for bright electron photo-injectors, high-gradient X-band structures at 12 GHz, and innovative short-period undulators. If compared to existing facilities, the proposed facility will (i) benefit from a lower electron beam energy, due to the enhanced undulator performance, (ii) be significantly more compact, as a consequence of the lower beam energy and the high gradient of the X-band structures, (iii) have a much lower electrical power demand and a smaller footprint. All of these enhancements will make our design more affordable to build and operate when compared against the existing facilities. The CompactLight prime objective is to generate a compact and affordable FEL facility design. Note that we aim to make the FEL affordable to operate as well as to build.

The specifications of this future FEL are driven by the demands of potential users and the associated science case. This report summarizes the findings of our interactions with potential users since the start of the design study through a number of different avenues, culminating in a dedicated CompactLight User Meeting that was held from the 27th to the 28th of November 2018 at the European Organisation for Nuclear Research (CERN) in Geneva, Switzerland [2]. The primary objective of the meeting was to consult potential users on the photon characteristics required by their current and future experiments. In addition to the dedicated User Meeting, two informal meetings with potential academic and industry users have been held in the UK, a specially developed questionnaire has been sent to over 50 FEL experts within Europe, and CompactLight have sent representatives to the Science@FELs Conference in Stockholm, Sweden in June 2018 and the Attosecond and FEL Science Conference in London, UK in July 2018, to hear about the latest scientific achievements using FEL

facilities and to informally interact with leading researchers to gather their views on the parameters and performance of future FELs.

2 Examples of groundbreaking research with x-ray FELs

2.1 Scattering

2.1.1 Bio-imaging

The structure of bio-molecules such as proteins, viruses or cells is essential for its function. Hence, the high-resolution structure determination enabled by coherent x-ray radiation is critical in the fields of biology and life science and permits e.g. rational drug design and the understanding of human biochemistry. A key method in this context is 'diffraction-before-destruction' in which the ultrashort duration of the x-ray pulse is exploited for outrunning the sample radiation damage. It offers the opportunity to image important bio-objects that can only be formed in smaller crystals, such as membrane proteins, or even single particles, with varying resolution. In particular, hard x-rays with wavelengths in the ångström range provide extremely high resolution [3] while soft x-rays offer useful information of larger structures, e.g. living cells, with high throughput. The ultrashort, intense x-ray pulses provided by FELs additionally allow measuring the dynamics of biologically relevant molecules on their natural, femtosecond, time scale.

Structure determination of micrometre-sized, or smaller, crystals at FELs is often carried out using serial femtosecond crystallography. Typically a liquid jet provides a stream of crystals that intersects the x-ray beam and the high intensity of the FEL x-ray beam enables the collection of diffraction images of thousands of randomly oriented crystals, which can be reconstructed into a 3D image with up to atomic resolution [4, 5]. Major progress with respect to the sample delivery has been made recently by increasing the speed of the liquid jet, which enables a fresh sample with every x-ray pulse at a MHz repetition rate [6, 7]. Simultaneous detector frame-rate development has also been carried out [8]. Another viable sample delivery method uses fixed targets and has important advantages such as an order of magnitude increase in the probability of hitting the sample by an x-ray pulse. This sample delivery method requires low kHz repetition rates.

A major scientific driver of x-ray FELs is the potential of single-particle imaging (SPI) of biological molecules at atomic resolution [9]. Although this goal is far from being reached there have been several measurements of biological objects at lower resolution. Moreover, SPI has tremendous potential for observing the dynamics of biological, chemical and physical systems [10]. Further progress requires development in sample delivery that optimizes the amount of useful collected data, specifically in aerosol injection which is advantageous in SPI because it provides a protective layer around the particle, thin enough to avoid excessive background scattering [11]. Recent studies [12] demonstrate aerosol particle speeds sufficient to cope with the 4.5 MHz intra-train repetition rate of the European XFEL.

2.1.2 Matter under extreme conditions

Intense laser pulses applied to solid materials can produce nanosecond or sub-nanosecond dynamic compression into extreme pressure regimes. The material response to the unexplored pressure and temperature conditions created by such compression can be uniquely explored in diffraction experiments with 100 fs temporal resolution that resolves the atomic motions. Moreover, the brilliance and small focus of the x-ray FEL pulse relaxes the pulse energy requirement of the optical pump laser, which can then also be focused to a small spot.

Studies of shock waves probed by 8-keV x-rays at FELs have measured the ultimate compressive strength, associated with a purely elastic response, of Copper, and the plastic flow occurring at higher strain orders [13]. Diffraction studies have also been applied to investigations of phase transitions and melting. For example, melting of Bi was observed after a few nanoseconds upon the release of the dynamically 8-14 GPa induced compression [14]. At LCLS shock pressures exceeding 120 GPa were used to demonstrate conversion of graphite to diamond and lonsdaleite phases [15] and complicated structures including linear guest structures arranged as chains in channels of the host structure have been observed. Moreover, pump-probe x-ray diffraction studies have revealed amorphous to crystalline transitions [16].

High energy-density plasmas are characterized by temperatures above 1 eV (~11600 K) and densities higher than that of a typical solid and are relevant in e.g. planet cores, stellar interiors, intense laser-matter interactions and fusion experiments. High-brilliance (hard-)x-ray FELs are well adapted for such studies as they satisfy both the requirements of generating and detecting the hot and dense plasmas deep into the sample. It also provides the necessary spatial resolution and the temporal resolution ranging from attosecond electron dynamics to compression processes on the nanosecond time scale. The potential of depositing mJ energies into a micrometre scale sample in less than 100 fs enables a homogeneous high-energy density distribution with known properties, which is formed faster than the hydrodynamic expansion. The first experimental demonstration of such an isochoric heating process was performed at LCLS in 2015 [17].

2.2 Spectroscopy

X-ray spectroscopy provides complementary information to imaging and diffraction measurements of the chemical and electronic properties of the system and the techniques are ultimately combined [18]. X-ray absorption (XAS) and emission spectroscopy (XES) enables element specific measurements of the unoccupied and occupied electronic states, respectively, while x-ray photoelectron spectroscopy (XPS) is highly sensitive to the chemical surroundings and offers surface sensitivity. The high-peak-brilliance FEL pulses allow new approaches to spectroscopic experiments such as nonlinear excitations, single-shot detection and femtosecond time-resolved measurements.

The potential for multiphoton excitation of atoms and molecules by FELs has been exploited for the formation of two-site double core-hole (tsDCH) states that could be detected using XPS [19, 20]. These states are created by the ejection of one core electron on separate atoms and require high peak intensities in order to ionize the second atom

before Auger decay occurs in the first atom. These states have received attention due to the significantly enhanced chemical sensitivity compared with single core-hole states.

XES and XAS studies carried out at x-ray FELs have been helpful in understanding important chemical processes. Electron transfer is essential in biological systems and for artificial light harvesting. XES and XAS measurements on an electron-harvesting chromophore performed at SACLA demonstrated the potential of these methods for monitoring fundamental chemical processes [21]. Moreover, resonant inelastic x-ray scattering has proven a capable tool for investigating excited state dynamics in solution via detection of orbital interaction [22]. The potential of using XAS and XES at FELs for understanding catalytic reactions have also been demonstrated at LCLS where these techniques have enabled the observation of CO oxidation on a Ru surface [23, 24].

X-ray magnetic circular dichroism (XMCD) spectra, obtained as the difference between XAS data with opposite circular polarization, offers a way to probe the magnetic properties of materials. Recently, the high peak brilliance of the soft-x-ray FEL was exploited in a demonstration of time-resolved (tr) XMCD, which was applied to investigate the element specific all-optical switching dynamics in GdFeCo with femtosecond temporal resolution [25]. Moreover, sub-picosecond demagnetization dynamics was studied by tr-XMCD using hard x-rays resonant with the Pt L_3 edge (11.6 keV) [26].

2.3 Time-resolved experiments

2.3.1 Ultrafast magnetism

Circularly polarized optical femtosecond pulses are able to reverse magnetization on a picosecond time scale [27]. X-ray FEL pulses offer an efficient tool for probing the ultrafast magnetization dynamics on a femtosecond time scale with nanometre resolution. For example, soft-x-ray holography using circular polarization has demonstrated 15 nm resolution [28]. Time-resolved soft-x-ray resonant diffraction studies carried out at LCLS detected Gd spin reversal within the first picosecond in the ferromagnetic GdFeCo. It was explained by a nanoscale flow of angular momentum from Fe-rich to Gd-rich regions induced by the optical pump [29]. The control of magnetic properties via the spins opens the door to faster data storage and processing devices.

2.3.2 Strongly correlated electron systems

In strongly correlated electron systems the interaction between electrons is non-negligible and may severely influence the character of the material. Light-induced insulator-metal transitions (IMT) dominantly driven by electron correlation effects as opposed to large structural changes, so-called Mott transitions, is a promising route towards faster electronics. Time-resolved x-ray diffraction experiments at FELs following the structural dynamics of the THz-induced IMT in VO_2 have shown that the electronic metallization dynamics and structural phase transitions can occur on different time scales [30]. This opens the door to efficient conductivity switching in correlated-electron systems.

X-ray FEL pulses are also useful for studying the lattice changes associated with light-induced superconductive phases. For example, THz pulses have been shown to

create superconducting properties in cuprate materials and using femtosecond x-ray diffraction the behavior of the lattice structure was investigated for this exotic state. These studies revealed that the nonlinear excitation of the crystal lattice structure creates a displaced lattice geometry which cause drastic changes in the electronic structure, and may cause destabilization of the charge-density wave order, of which both may favour superconductivity [31].

2.3.3 Water dynamics

Water is a surprisingly complex liquid that is still far from understood. The reason for its complexity and anomalous properties is the ability to form highly disordered hydrogen-bonded networks. X-ray FELs permit resolving the water structural dynamics on a sub-100-fs time scale and atomic length scales. In a recent experiment at LCLS [32] using 8.2 keV photon energy water structural motions was observed from the decay of speckle contrast when tuning the pulse duration from 10 to 120 fs. They showed that cage effects due to hydrogen bonding play an important role in the slower dynamics of water upon cooling.

3 Science requirements on a next-generation FEL

3.1 Trends in the FEL science community

Representatives from CompactLight participated in and presented posters at two conferences devoted to science research at FELs: (i) Science@FELs Conference in Stockholm, Sweden in June 2018 and (ii) Attosecond and FEL Science Conference in London, UK in July 2018. The presented talks and informal discussions with FEL users showed trends towards experiments demanding higher photon pulse energies, better focus and shorter pulse durations. For instance, there is an increased interest in using FELs to explore:

1. materials far from equilibrium such as light-induced superconductivity;
2. nonlinear X-ray optics;
3. multidimensional attosecond spectroscopy;
4. charge migration and ultrafast x-ray damage in biomolecules;
5. surface chemistry and pathways for catalysis;
6. matter under extreme conditions.

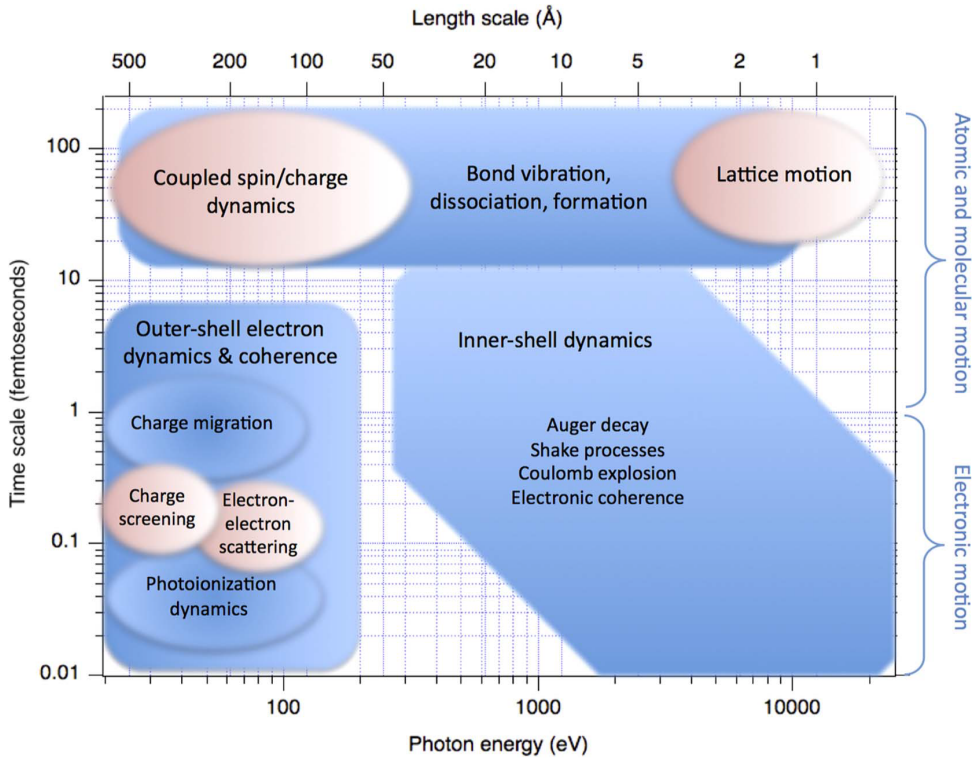

Figure 1: Characteristic time and energy scales of fundamental processes in atomic, molecular, electronic, spin and lattice systems. The characteristic length scales are indicated on the top bar. The minimum time and length resolutions for a given photon energy are limited by the Heisenberg uncertainty.

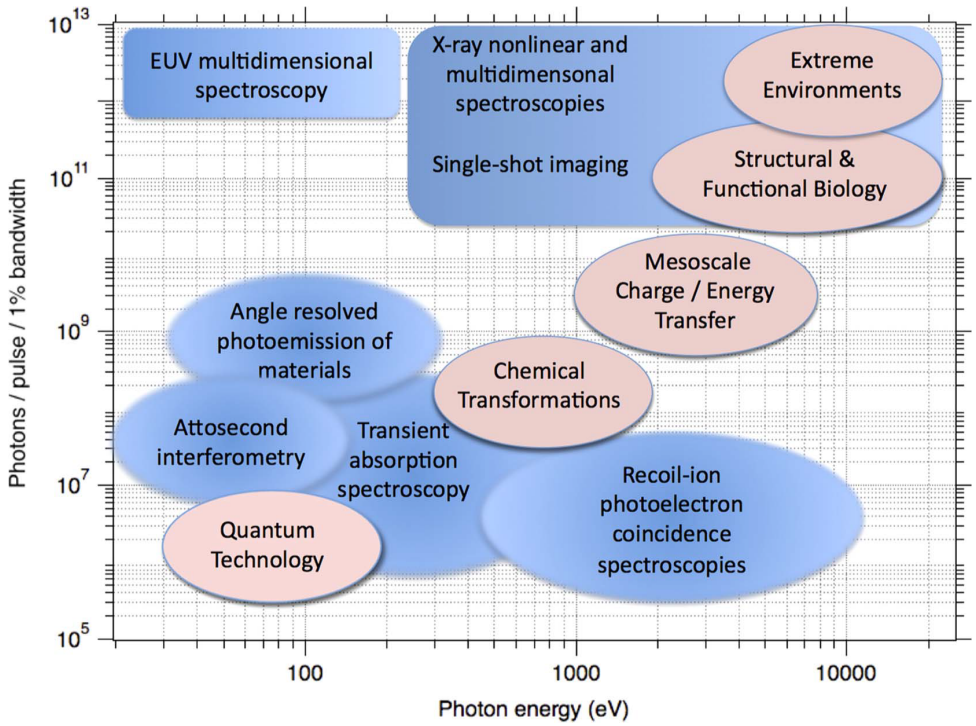

Figure 2: Number of photons per pulse into 1% bandwidth as required by different experimental ultrafast x-ray techniques (blue). The research areas relaying on the techniques are shown in pink. The high-fluence regime enables nonlinear x-ray spectroscopies and single-shot imaging, potentially with atomic resolution. Figure adopted from Ref. [33].

The trends towards shorter pulse durations and higher photon pulse energies has been recently reviewed by a group of users from around 20 universities and national laboratories worldwide [33]. The photon requirements for different disciplines are graphically presented in Figs. 1 and 2. The requests for next-generation FELs collected in informal discussion at the conferences fully support the data presented in the figures. In addition, during the discussions there were very strong requests for improving the coherence and stability properties of FEL radiation pulses as well as much better synchronization to external laser sources.

3.2 Survey on user requirements

In conjunction with the CompactLight User Meeting held at CERN, a preliminary survey was conducted through the use of an online questionnaire. The purpose was to gather quantitative information about the user requirements on the photon characteristics. A total of 10 responses were received from potential users.

The respondents have expressed interests in experiments such as (i) pump-probe diffraction, (ii) serial crystallography, (iii) time-resolved spectroscopy and (iv) time-resolved scattering. In addition, each respondent has specified either one or two sets of desired parameter values for the future x-ray FEL. These parameter values are shown as histograms in Fig. 3.

With regard to the tunability, there is a clear demand for photon energies as low as

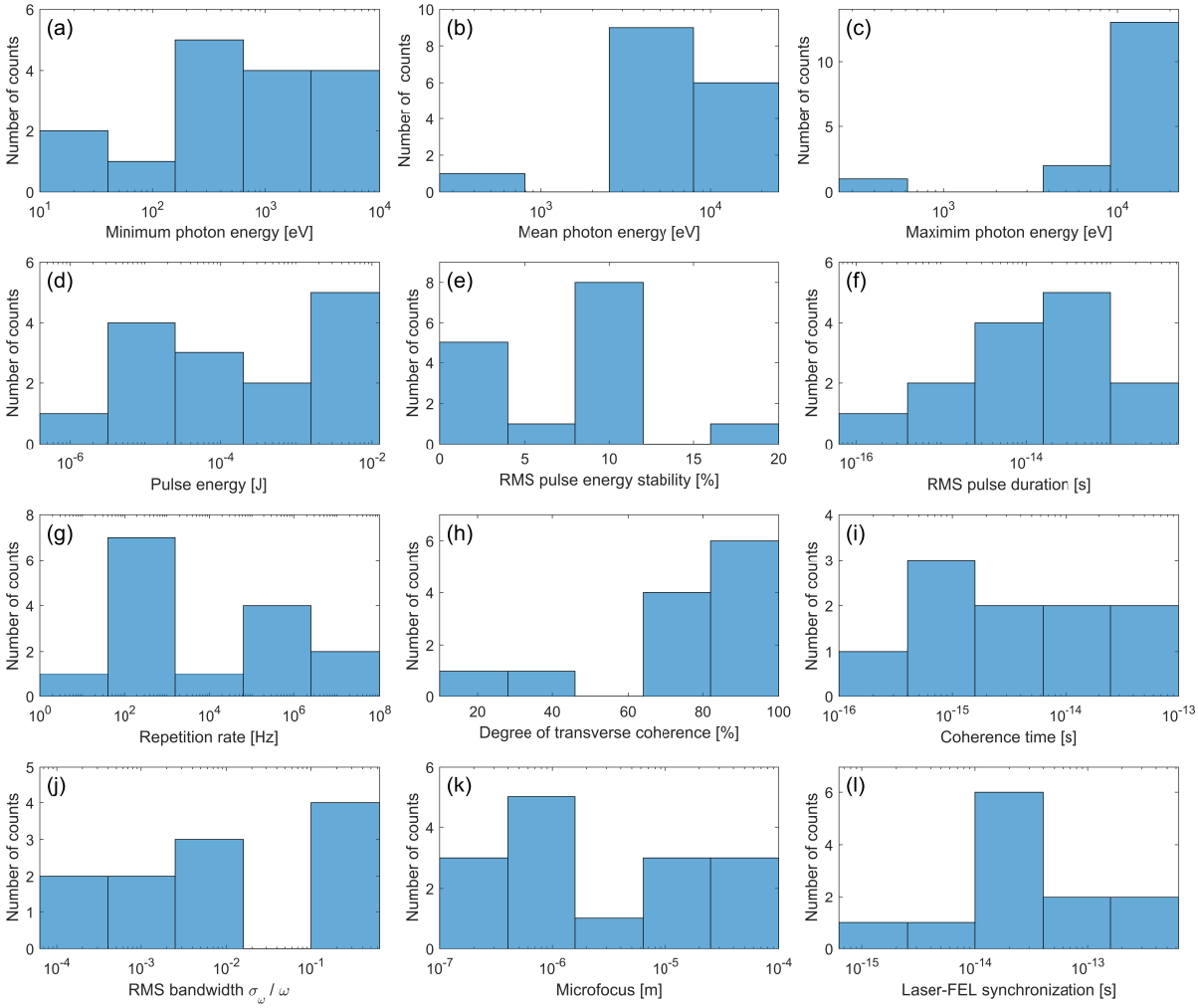

Figure 3: The results of the survey are summarized in histograms showing the users' requirements on the (a) minimum photon energy, (b) mean photon energy, (c) maximum photon energy, (d) pulse energy, (e) pulse energy stability, (f) pulse duration, (g) repetition rate, (h) transverse coherence, (i) longitudinal coherence, (j) bandwidth, (k) focused spot size and (l) synchronization between the FEL and the external laser.

0.2 keV [see Fig. 3(a)] and as high as 20 keV [see Fig. 3(c)]. The mean photon energy of the desired tunable range is about 4 keV [see Fig. 3(b)].

The preferable pulse energy is in the range of 3–100 μJ [see Fig. 3(d)]. Furthermore, the demand on the *stability* of the pulse energy is stringent, and most respondents want the RMS fluctuation in pulse energy to stay below 12% [see Fig. 3(e)].

Most respondents prefer a pulse duration of 10–100 fs [see Fig. 3(f)], a repetition rate higher than 100 Hz [see Fig. 3(g)], a degree of transverse coherence higher than 70% [see Fig. 3(h)], a coherence time of 1–100 fs [see Fig. 3(i)], a bandwidth of 0.1–1% [see Fig. 3(j)] and a microfocus of 0.1–100 μm [see Fig. 3(k)]. For pump-probe experiments, most respondents want the synchronization between the FEL and the external laser to be in the order of 10 fs [see Fig. 3(l)].

The questionnaire also asks the potential users to comment on any FEL feature that would benefit their future experiments. The answers are summarized as follows:

- variable polarization (linear and elliptical);

- pulse energy above 3 mJ;
- shorter pulse duration;
- higher stability in pulse energy and pulse duration;
- repetition rate of 1–10 kHz;
- laser-FEL synchronization better than 50 fs;
- FEL-pump FEL-probe capabilities with a large photon energy difference;
- small focused spot size;
- tunability extended to higher photon energies;
- better reliability of two-colour pulse generation.

4 Summary of Findings and CompactLight Specification

This report summarizes all of the discussions and interactions that the CompactLight collaboration has held with potential users of the facility during the past year. The exploitation of FELs by numerous groups covering a diverse range of research topics highlights why FELs are incredibly important engines of discovery. The diversity also means that it is not possible to meet the current and future needs of all users with a single facility. Indeed, there is a risk that by trying to satisfy all requirements the facility performance would be compromised and no users would be entirely satisfied. The CompactLight collaboration understands this issue and so has distilled all of the user input into a coherent specification that is fully aligned with our prime strategic objective which is to generate a compact and affordable FEL facility design.

The specification of the CompactLight FEL is summarized in the following bullet points and in Table 1.

- Stability in all its aspects is very important to all experiments. We will bear this in mind in all of our technical designs of systems and sub-systems.
- Seeding of the FEL enhances the output quality significantly and will be implemented at all wavelengths, where feasible, and where compatible with our compact and low-cost objective.

Table 1: Main parameters of the CompactLight FEL.

Parameter	**Unit**	**Soft-x-ray FEL**	**Hard-x-ray FEL**
Photon energy	keV	0.25 – 2.0	2.0 – 16.0
Wavelength	nm	5.0 – 0.6	0.6 – 0.08
Repetition rate	Hz	1000	100
Pulse duration	fs	0.1 – 50	1 – 50
Polarization		Variable, selectable	Variable, selectable
Two-pulse delay	fs	±100	±100
Two-colour separation	%	20	10
Synchronization	fs	<10	<10

- Peak Brightness is key to many experiments and we will aim to maximize this where compatible with our compact and low cost objective.
- Extreme synchronization between different photon sources for time-resolved pump-probe experiments is vital and we will provide a design that can synchronize the FEL with a conventional laser to better than 10 fs.
- Two pulses and two wavelengths are essential for many experiments. We will develop a design that provides these capabilities in as compact and low cost a way as possible.
- Generating pulses as short as 100 attoseconds is desirable but may take significant extra space and cost. In this case we will relax the specification to a point where the cost impact is negligible.
- A repetition rate of 1000 Hz for the soft-x-ray FEL will be a unique and highly desirable feature of our facility. We recognize that this is a very challenging target for many systems and that we may have to compromise on this aspirational target during the course of the Design Study.
- The photon pulse bandwidth will be minimized to maximize peak brightness where compatible with our compact and low cost objective.
- The facility output will cover the range between 250 eV and 16.0 keV with all photon energies within this range being accessible from at least one of the FEL beamlines.
- The 2 keV "boundary" between the soft-x-ray FEL and the hard-x-ray FEL is not rigid and will be determined by the collaboration when considering all the technical options including electron beam energies, undulator performance, and x-ray optics capabilities.
- Tuning across photon energies will primarily be achieved by undulator scanning rather than energy scanning to maximize the efficient operation of the facility. We will operate the FELs at a few discrete electron beam energies, as required, to achieve the full wavelength tuning ranges.
- The FEL output pulses will be evenly spaced in time and not provided in a burst mode.
- To maximize the efficient use of the facility we recognize that simultaneous operation of both the soft- and hard-x-ray FELs would be beneficial. We will investigate options for achieving this where compatible with our compact and low cost objective.
- We recognize that output pulse energies will naturally be reduced when we operate at the very shortest pulse lengths, as they do at other FEL facilities. We will ensure that our pulses remain competitive with these other facilities.
- Variable, selectable polarization is required at the sample for all photon energies. We will investigate options for the production of such variable polarization at different photon energies using either optical elements in the photon beamline or elliptical undulators.

The target performance of CompactLight in terms of peak brilliance is shown graphically in Fig. 4. The peak brilliance is expected to be comparable to the state-of-the-art x-ray FEL facilities which are currently in operation.

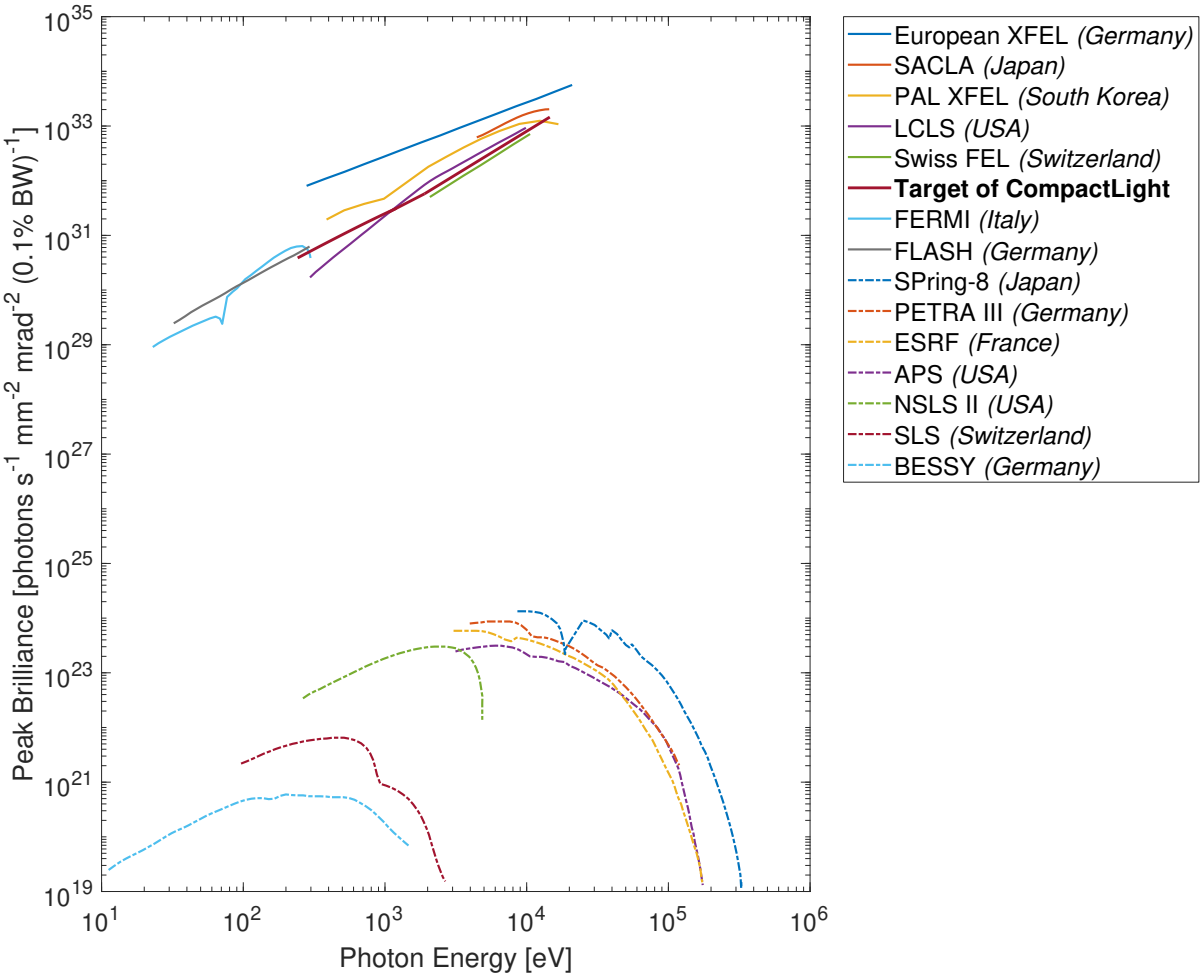

Figure 4: Peak brilliance as a function of photon energy for a selected set of x-ray sources. Free-electron laser facilities are shown in solid lines, and synchrotron facilities are shown in dashed lines. This figure is adapted from Fig. 1 in Ref. [34].

Appendix A Responses to online questionnaire

As discussed in Section 3.2, a preliminary survey was conducted by means of an online questionnaire in conjunction with the CompactLight User Meeting held at CERN. The purpose was to gather quantitative information about the user requirements on the photon characteristics. In each of the 10 responses received from potential users, either one or two sets of desired parameter values for the CompactLight FEL were specified. These parameter values are listed in Table 2.

Table 2: Photon characteristics specified by potential users in the online questionnaire. Blue columns correspond to diffraction or scattering experiments. Yellow columns correspond to spectroscopy. Orange columns correspond to interactions between x-ray and matter. Grey columns correspond to other experiments.

Min. photon energy [keV]	4.5	0.05	0.01	3	1	0.5	0.5	0.01	1	0.2	3	3	0.7	0.5	2	0.2
Max. photon energy [keV]	12.6	7	10	20	12	10	15	0.5	9	10	16	16	10	10	20	20
Repetition rate [Hz]	120	1000	100000	1.1		100	1000	100000000	120	1000000	1000000	4500000	100	10000	1000	1000000
Pulse energy [μJ]	100	250	100	1000	2000	1	5		3000	10000	2000	5000	10	10	10	100
RMS pulse energy stability [%]	20	10	10	0.1	1	10	1		10	10	10	10	10	0.1	0.01	5
Microfocus [μm]	1	1	0.5	10	0.1	10	5		1	0.1	1	0.25	100	100	10	30
Degree of transverse coherence [%]	100	80	100	80			100			90	80	80	100	100	30	10
Coherence time [fs]	1	2	0.2	1							2	10	1	100	50	10
RMS bandwidth [%]	0.05	20	10			0.01	0.01		0.3	0.3	0.5	0.2			10	10
FWHM pulse duration [fs]	10	2	0.2	50		40	10		60	1	50	10	50	10	100	500
Two-pulse spectral separation [nm]	0.6												0	0	100	100
Two-pulse temporal separation [fs]	100	150	10										0	100	10000	1000
Laser-FEL sync [fs]	10	10	1	300		40	10				200	30	50	10	10	5

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