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New generation of light sources: Present and future☆

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ABSTRACT

Spectroscopy and imaging in the VUV–X-ray domain are very sensitive tools for the investigation of the properties of matter [1–3]. Time-resolved studies enable to follow the movies of ultra-fast reactions. More than fifty years after the laser discovery [4], VUVX light sources are actively developed around the world. Among them, high order harmonics generated in gas, X-ray lasers, synchrotron radiation, free electron lasers are providing a wide offer, from laboratory size sources to large scale facilities, with various features, suitable for different types of experiments. The properties of these sources are here reviewed. Quest of new performances and flexibility is also discussed.

1. Introduction

Different types of light sources for the investigation of matter [1–3] are suitable for applications in the VUV-X domain. The X-ray laser and high order harmonic generation in gas (HHG) [5,6] or on solid targets [7] take advantage of the light emission properties of matter. Recently, the use of single, few-cycle laser pulses in the mid-infrared enabled to provide tunable radiation to above keV energies with a few attosecond duration [8]. Synchrotron radiation from third generation light sources and free electron lasers (FEL) [9] rely on synchrotron radiation generated from charged particles in bending magnets or undulators, creating a periodic permanent magnetic field [10]. Partial transverse coherence is provided by the low value of the emittance (product of the beam size by its divergence) and a quest for ultimate storage rings is now under way for improving the coherence for imaging applications. The longitudinal coherence is achieved in FELs [11] by setting in phase the electrons, thanks to an energy exchange between the electrons and a light wave (the spontaneous emission or an external seed) resulting in bunching. Presently, LCLS (Stanford, USA) [12] and SACLA (Harima, Japan) [13] are tunable, femtosecond X-ray sources in the 1–0.1 nm spectral range, with up to several mJ energies, operating in the self amplified spontaneous emission regime. In complement to these, FLASH [14], the SCSS test accelerator [15] and FERMI [16] are operating in the XUV region, FERMI being the first seeded FEL open for users. HHG is also used as a seed for FEL or X-ray lasers [17]. Besides, laser wakefield acceleration [18] is rapidly developing and it is already possible to generate synchrotron radiation (the so-called betatron radiation [19]) thanks to the ion field, or by Thomson scattering [20]. One can further consider setting up a free electron laser using these electron beams, provided an adequate beam manipulation through the transport to the undulator [21]. Future developments of these various sources, including interplay between them, target now to deliver radiation with more versatile properties, and will be briefly discussed.

2. HHG and X-ray lasers

High order harmonic generation in gas appeared at the end of the eighties [5,6]. An intense laser beam is focused in a rare gas (such as Xe, Ar, Ne...) from a cell of a jet producing a harmonic beam consisting of odd harmonics. The strong laser field enables tunnel ionization of the electrons, which are then accelerated and diffused in the atomic potential, and recombine in emitting harmonics. The cut-off energy is determined by the ionization potential of the atoms (21.6 eV for Ne, 24.6 eV for He), and by the ponderomotive energy scaling as the square of the laser wavelength and decreasing for longer pulse durations.

Besides the change of wavelength by stepping from one harmonic to another, further tuneability has first been achieved by taking advantage of the laser tuneability itself, achieving full tuneability from 220 nm to 8 nm with 1.1 to 1.6 μm pump laser (Ti:Sa laser coupled to an optical parametric amplifier) [22], ~70% tuneability from 180 to 18 nm by frequency mixing [23], by adjustment of the laser energy and chirp [24]. Further tuneability is later produced using long wavelength laser and few cycle pulses, as described further.

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The HHG wavelength has been decreased by steps. Water window has been reached in the end of the nineties, with 2.7 nm (460 eV) in He on the 221st harmonic, and with 5.2 nm (239 eV) in Ne from a 800 nm, 26 fs laser [25]. 7.75 nm (160 eV) has been produced in Ar with an OPA [26]. Then, 0.95 nm (1.3 keV) has been delivered from a 720 nm, 5 fs, 0.2 TW laser at 1 kHz in He [27]. Phase matching enabled then to raise the HHG intensity at short wavelength, at 4.4 nm with quasi phase matching by periodically modulating the diameter of a gas filled hollow waveguide with a 1 kHz, 22 fs, 1–3 mJ Ti–Sa laser [28], and at 4.37 nm with 10 nJ/pulse with quasi phase matching in a Ar capillary [29].

In the temporal domain, HHG usually generated attosecond trains in a femtosecond envelope [30]. Attosecond pulses result from the interference of the back-driven wavepacket by the intense laser field after ionization with the bound one. The attosecond pulses can be controlled by temporally confining the photons to one single burst, according to the phase of the laser electric field. A 250 as control has been achieved with a 5 fs, 0.5 mJ, 1 kHz, 750 nm linearly polarized laser in a 2 mm Ne gas medium [31]. Then, 80 attosecond pulses have been generated in the XUV at 110 eV with a 3.3 fs (1.5 cycle), 720 nm linearly polariszed laser, with two subsequent jets of Ne atoms gas, the second one being use for the detection [32]. Isolated attosecond pulses can also result from polarization gating, in taking advantage from the strong HHG sensitivity to the ellipticity of the fundamental field (maximum for linear polarization). A laser that is only linearly polarized for a short time (otherwise elliptically polarized) enables to temporally confine the emission. Attosecond pulses have been isolated in such a way using a carrier envelop phase (CEP) – stabilized – 5 fs pulse, combined with birefringent plates producing a polarization gate shorter than half a laser period [33].

Cut-off energy has been extended using mid infra-red driving lasers and shorter pulse drivers. Indeed, 7.75 nm (160 eV) has been reached in Xe using a 0.8–1.8 μm CEP laser with 11–73 fs pulse duration [34]. Harmonics being very close one to each other, a quasi-continuum is emitted. A striking enhancement around 95 eV results from the giant resonance in xenon (multi-electron correlation upon the recombination step in HHG). Moreover, in the temporal domain, near single attosecond pulses are achieved. Furthermore, radiation is produced beyond the nanometer, with 0.77 nm (1.6 keV) generated in high pressure helium with a 6 cycle 0.8–3.9 μm [35] with phase matching. In such a case, the Fourier transform of the measured spectum suggest that the X-ray pulse would last only 2.5 attoseconds, provided the chirp is compensated.

HHG present a high level of transverse coherence, as demonstrated by various measurements such as Fresnel bi-interferometer [36], by Young slits [37,38]. For example, HHG can offer a divergence of 0.35 mrad with pulses of 25 nJ at 13.5 nm with Ne gas, and of 0.3 mrad with pulses of 1 nJ at 8.9 nm in He [39].

HHG have been usually produced with standard laser, without specific effort toward high repetition rate. However, some measurements have been carried out with 1 kHz Ti–Sa lasers, such as 5.10^{10} ph/pulse at 20–32 nm in Ar with a 5 mJ, 1 kHz laser [40], 10^{10} ph/pulse (50 nJ) at 20–32 nm in Argon with a 7 mJ, 1 kHz [41], or 90 nJ at 30 nm [42]. Intra-cavity HHG enables to reach the MHz repetition rate, with typically 100 nW [43]. Fiber lasers have also been used, providing 7.9×10^{11} ph/pulse at 17 nm, with a 100 kHz, 100 μJ laser [44]. Developments are under way for enhancing the fiber laser repetition rate and output power, by cavity enhancement [45], by phase locking of fiber amplifiers [46], enabling to expect a bright future of fiber lasers [47].

HHG generally presents a linear polarization. However, elliptical polarization can be produced in HHG from aligned molecules, resulting from the phase difference between the parallel and perpendicular components of the dipole (due to non-isotropic Coulomb potential, the recolliding electronic wave differs from a plane one, leading to multiple orbital dynamics) [48,49] or to multiple orbital dynamics [211].

Efforts have been taken to improve the HHG efficiency, by modifying the atomic response by changing the driving electric field (and thus by breaking the symmetry between consecutive half cycles). The addition of a second harmonic field enables to generate both even and odd harmonics [50,51]. The addition of the third harmonic field leads to an efficiency increase by a factor 10 [52]. The addition of the below threshold low order harmonics (10 nJ) in a dual gas cell configuration leads to a HHG energy increase and to a larger spectral range [53]. The last configuration can also be viewed as a HHG seeded HHG.

High order harmonics can also be generated from solid targets. An intense laser pulse interacts with a near discontinuous plasma-vacuum boundary so that the laser electric field can efficiently couple to the plasma surface. In consequence, the electrons oscillate in phase, acting as a relativistic mirror oscillating at the laser frequency. A temporal function of the incident optical laser cycle corresponds to the position of this mirror surface. The phase of the reflected light wave is modulated, it becomes no longer sinusoidal and leads to the emission of a high order harmonics content. Radiation up to 3.3 Å (3.8 KeV) on the 3200th harmonics has been efficiently produced [54–56].

X-ray lasers, relying on the population inversion by electron ion collision in hot highly ionized plasma operate in the amplification of spontaneous emission (ASE) mode [57]. Step by step tuneability is achieved, pulse duration is typically of 100 ps duration [58]. Seeding can significantly improve the performances of these X-ray lasers: HHG seeding enables to sharpen the divergence, enhance the emitted intensity [59], whereas seeding with the free electron laser leads to a 1.46 nm inner-shell X-ray laser [60].

3. Synchrotron radiation

Synchrotron radiation is the electromagnetic radiation emitted by accelerated charged particles. After the establishment of the first theoretical foundations [61,62], analysis was carried on angular and spectral distribution and polarization properties [63], on the influence of the energy losses due to radiating electrons on the limit on the obtainable energy in a betatron [64]. The synchronism for the particle loosing energy by synchrotron radiation can be maintained via the injection of the particle bunch in proper radio frequency phase as proposed theoretically [65] and shown experimentally [66]. The spectrum distribution was described in [67]. The first synchrotron radiation was then observed in the visible, tangent to the electron orbit one year later on the 70 MeV general electric synchrotron, of 29.3 m radius and 0.8 T peak magnetic field [68]. The rapid increase of the intensity with the electron beam energy was measured (fourth power of the energy). The emitted light was found polarized with an electron vector parallel to the plane of the electron orbit. The radiation from a relativistic particle in the magnetic sinusoidal field has also be analyzed [69] and observed [70]. In a storage ring, the energy loss due to synchrotron radiation is compensated thanks to a radio frequency field.

Synchrotron radiation is produced when the particle trajectory is subjected to a magnetic field, which is for example generated in bending magnets in circular accelerators. Thanks to the relativistic projection of angles the radiation is collimated in a thin cone which angle corresponds typically to the inverse of the normalized energy γ of the particles, the higher the electron beam energy, the higher the collimation. Synchrotron radiation covers a wide spectral range and can be tuned from the infra-red to the X-rays.

It can also be produced in the so-called insertion devices, undulators and wigglers, which create a periodic permanent magnetic field (amplitude B_0, period λ_0). In the case of an undulator creating

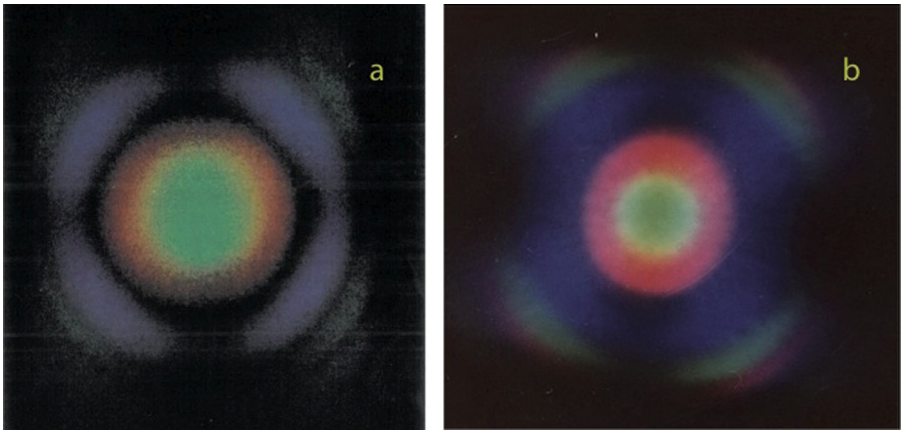

Fig. 1. ACO undulator radiation pattern for two different undulator gaps corresponding to two a central wavelength. The off axis emission consists of radius of wavelength larger than the resonant one, thank to $\gamma^2\theta^2$ the term. Red rings correspond to the off-axis emission of UV harmonics. (For interpretation of the references to color in this text, the reader is referred to the web version of the article.)

a sinusoidal field in the vertical plane, the synchrotron radiation on the axis is emitted at the wavelength λ, so-called resonance wavelength, and its odd harmonics of order n, with a linear horizontal polarization: $\lambda = \lambda_0(1 + K^2/2 + \gamma^2\theta^2)/2n\gamma^2$, with K the deflexion parameter of the undulator, $K = 0.94\,\lambda_0(\mathrm{cm})\,B_0(T)$ and θ the observation angle. Fig. 1 shows a radiation image pattern from an undulator.

In the "undulator" regime (rather small K value), the radiation emitted at each inversion interferes with the one produced in the previous inversions. These interferences can be constructive and the radiation is produced in a very intense spectral rays (harmonics) form. The sharpness of the harmonics can be affected by the observation angle, the energy deviation of the particles (energy spread) and the spatial and angular extension of the electron beam (with the so-called "emittance" contribution with the beam emittance defined as the product of its transversal dimension by its divergence). In the "wiggler" regime ($K \gg 10$), the radiation of the different harmonics overlaps and is similar to the dipole one, with a higher intensity. Fig. 2 presents an example of the influence of the contribution of the electron beam energy spread and emittance on the undualtor harmonic linewidth. For a mono-energetic filament beam and odd harmonics emitting on axis, the nth harmonics bandwidth is given by $1/nN$ with N the number of undulator periods. Sharp lines result from large number of periods, and high harmonic number. The electron beam emittance enlarges the undulator line in its red part, since the emission of non on-axis electrons can be considered off-axis, a sideband can even appear. The energy spread enlarges symmetrically the undulator line. Even harmonics are appearing on-axis with the emittance and energy spread contribution. Synchrotron radiation is generally used after a high resolution monochromator, for selecting the proper bandwidth of analysis.

The wavelength λ of the emitted radiation can be varied by a modification of the undulator magnetic field (by changing the gap for permanent magnet insertion devices or the power supply current for electromagnetic insertion devices). The particular choice of the undulator characteristics and technology enables to optimize the desired spectral range for a given beamline. Fig. 3

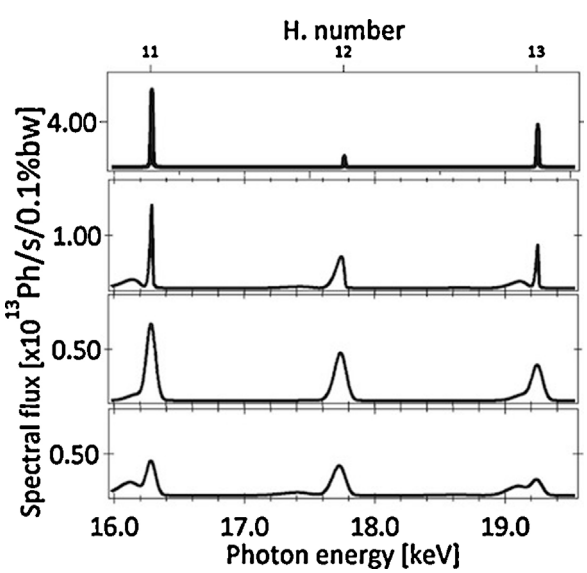

Fig. 2. Undulator harmonic bandwidth dependence on the electron beam characteristics: up: case of a filament mono-energetic electron beam, below: with the contribution of the emittance (horizontal emittance of 3.9 nm rad and vertical emittance of 39 pm rad), below: contribution of energy spread (0.1%), down: joint contribution of emittance and energy spread. SRW calculation in the case a U20 SOLEIL in vacuum undulator at minimum gap (5.5 mm) at 2.75 GeV.

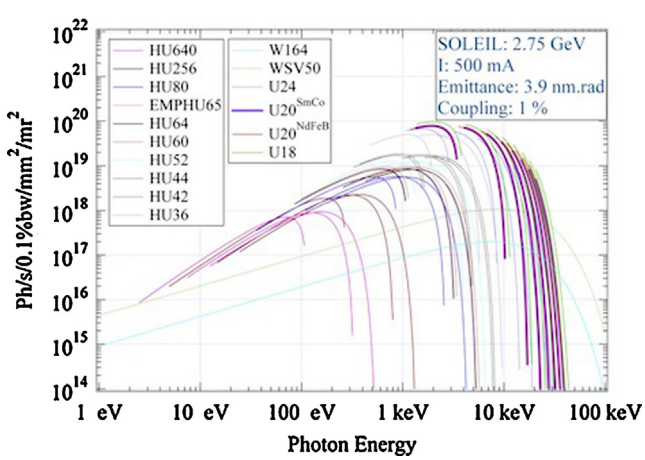

Fig. 3. SOLEIL synchrotron radiation spectral range for the various insertion devices: electromagnetic undulators of 640 mm and 256 mm periods (HU640, HU256) for the long wavelength region, various elliptically polarized undulators for the VUV and soft X-ray region (with periods from 80 to 36 mm: HU80 to HU36), in-vacuum undulators for radiation at several keV on the undulator harmonics (U20, U24) with shorter periods (20, 24 mm), cryogenic in-vacuum undulator operating at 77 K for higher remanent field and coercivity (U18), in-vacuum wiggler for extension to higher energies (WSV 50 of 50 mm period) and an out of-vacuum wiggler for 164 mm period (W164). SRW calculations performed with an emittance of 3.9 nm rad, 1% coupling, 0.1% energy spread.

Fig. 4. Synchrotron radiation sources around the world.

presents the spectra calculated with SRW [71] for the different insertion devices of SOLEIL 2.75 GeV synchrotron light source in France. In-vacuum undulators [72] enable to reach a higher field by placing directly the magnets inside the vacuum chamber. By cooling down $Nd_2Fe_{14}B$ permanent magnets, B_r is increased by 10% and the coercivity by a factor of 3 [73,74]. $Nd_2Fe_{14}B$ base cryogenic undulators [75] should be operated around 130–140 K because of the spin reorientation transition occurring at lower temperatures [76] requiring the cryogenic undulator to be cooled down to the liquid nitrogen temperature and heated back to the working temperature to 140 K. $Pr_2Fe_{14}B$ based cryogenic undulators [77] can be directly cooled and operated at 77 K. Superconducting undulators, for which active R&D is under way, can potentially lead to high magnetic fields [78–81]. For the spectral range extension toward higher photon energies, in-vacuum wigglers [82–84] offer a good alternative to superconducting wigglers [85] with an easier daily operation.

The electric field of the radiation is in the plane of the electron trajectory. For a vertical magnetic field, the electron follows a wiggler trajectory in the horizontal plane. Combining magnetic fields in both planes with a possible phasing between them enable to provide various type of polarization from liner vertical, linear horizontal to circular one, or more generally elliptical one. Electromagnetic technology with [86] or without poles [87] suits well for the fabrication of rather long period elliptically polarized undulators (EPU), providing the possibility of any type of polarization. Permanent magnet based schemes also exist, such as crossed undulators [88], HELIOS [89], Diviacco/Walker scheme [90], APPLE-I [91], APPLE-II [92], APPLE-III [93], a 6-arrays device [94], DELTA [95]. Combining electromagnets and permanent magnet provides a fast switching of the polarization from circular right to circular left and vice versa [96–98].

The storage ring radio-frequency cavities re-accelerate the particles at each turn and give them back the energy they have lost by synchrotron radiation. The electrons are packed in a large number of bunches with a bunch length of about ten picoseconds (FWHM), being imposed by the dynamic equilibrium linked to the high number of turns. The electron longitudinal oscillation imposed by the radio-frequency system induces a pulse minimum duration of few tens of picoseconds, to which adds a systematic bunch lengthening following the interaction with the emitted microwave field [99]. There are various strategies for shortening the radiated pulse duration e.g. [100]. The simplest one is to operate with a specific electron optics (low momentum compaction factor) providing short pulses at low current [101]. Another strategy is to flip the transverse phase space to the longitudinal one and vice versa. Otherwise, the "so-called" slicing techniques using the interaction of a femtosecond laser with the electron bunches in an undulator [102] can produce subpicosecond radiation pulses on these rings, but to the detriment of the total flux. Different simultaneous short bunches can also be achieved depending on the phase of different superimposed RF fields [103].

Spatially coherent synchrotron radiation from storage rings is achieved for an electron beam emittance of the order of the targeted wavelength. The emittance depends on the lattice, the bending angle per dipole, the partition number, and of the dipole and wiggler energy loss [104]. There are different strategies for emittance reduction [105] for the so-called ultimate storage rings, such as the use of the high number of dipoles and of the minimization of the lattice function with strong focusing and multibends, the use of damping wiggler (which leads however to an increase of the energy spread), and by a change of the partition number, in using a Robinson wiggler for example [106]. For example, PETRA III at 6 GeV [107] reaches 1 nm with a large circumference and the use of damping wigglers, NSLS-II at 3 GeV [108] is aiming at 0.5 nm, MAX-IV at 3 GeV aims at 0.24 nm with a 7 bend achromat optics and the use of damping wigglers [109], ESRF at 6 GeV targets an emittance reduction in two step, from 4 nm rad to 0.15 nm rad and 0.01 nm rad, corresponding to an increase of the undulator brightness (the number of photon per cell in phase space) by a factor 25 and then 5 [110].

Synchrotron radiation light sources are widely developed around the world, as illustrated in Fig. 4.

The common approach is to define the brilliance as the number of photons per second and per unit of phase space (or the density distribution in phase space), which corresponds to the geometrical optics frame. It is defined in the general case in the frame of the Wigner distribution [111,112]. It can be approximated in the case

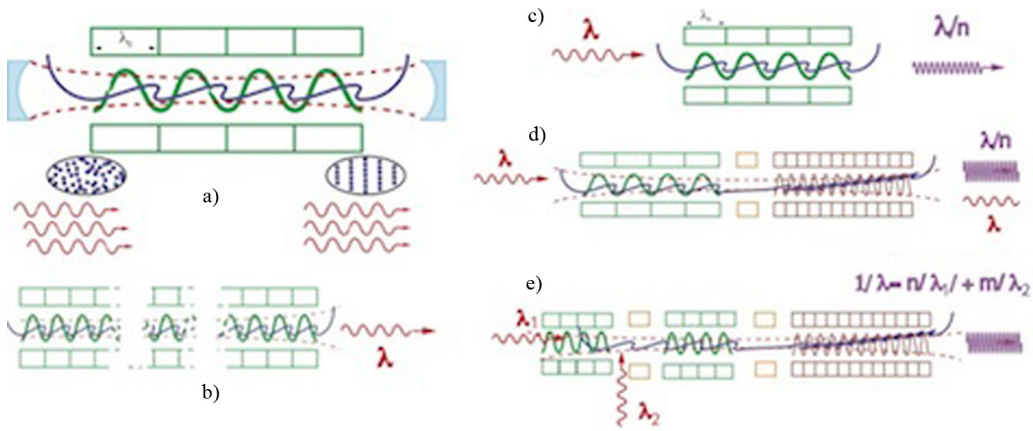

Fig. 5. Free electron laser configurations: (a) oscillator case with an optical cavity enabling to store the spontaneous emission, (b) self amplified spontaneous emission (SASE) where the spontaneous emission emitted in the beginning of the undulator is amplified in one single pass, (c) seeding where a coherent source tuned on the resonant wavelength of the undulator enables to perform efficiently the energy exchange leading further to the density modulation, (d) high gain harmonic generation, and (e) echo enable harmonic generation (EEHG).

of Gaussian beams [113]. The Wigner distribution approach of the brilliance concept inherently incorporates the complete information on the electric field, from different position. It thus properly provides information on the transverse coherence [114]. Similarly, there is some link between peak brilliance and longitudinal coherence.

Ultimate storage rings with low emittance aim being at the diffraction limit. Concerning longitudinal coherence (light intensity scaling as the square of the number of electrons), it occurs when the electron bunch is shorter than the considered wavelength, typically in the THz spectral region on current storage rings [115] or if a micro-bunching of the electron takes place, such as in the free electron laser (FEL) process for example.

4. Free electron lasers

With a FEL (see Fig. 5), the generated radiation is not only based on the spontaneous synchrotron radiation emitted in the undulator: a light wave of wavelength λ interacts with the electron bunch in the undulator, inducing an energy modulation of the electrons; which is gradually transformed into density modulation at λ, the phased electrons then emit coherently emission at λ and its harmonics of order n. The consequent light wave–electron interaction leads, in certain conditions, to a light amplification to the detriment of the kinetic energy of the electrons. The small signal gain is proportional to the electronic density and varies as $1/\gamma^3$, depending on the undulator length. Saturation comes from the fact that the resonance condition by the light and the radiation from the undulator is no more fulfilled, the electron having lost too much energy, or from an increase of energy spread [116,117]. In addition, the light traveling slightly quicker than the electrons (slippage), the light slips over one wavelength for one undulator period (slippage); and it could also escape from the electron bunch for short ones and quite long undulators.

FELs can be implemented on different types of accelerators for providing the electron beam. Storage rings provide rather long electron bunches (10–30 ps) because of the electron beam recirculation and the emittance scales as the square of the electron beam energy. Linear accelerators, single pass machines, provide quite short bunch 10 fs–10 ps duration, of interest for ultra short pulse source production and for high electron beam densities, emittance also scales as the inverse of the energy, enabling to reach diffraction limit sources for high electron beam energies required for short wavelength operation. Energy recovery linac combines both advantages of the two previous accelerator types, with short pulses, few turn recirculation and energy recovery for power consumption saving. For both linac based FELs and ERL, RF photo-injectors are currently used. The quest of low emittance, short bunches, high current can lead to conditions where the space charge [118] effects are limiting. Besides from the elementary process studies [119], RF gun studies indicate that an uniform photo-electron distribution from the cathode contained in an ellipsoid is optimum in the case of a space charge dominated beam, since forces are linear and the emittance growth induced by those forces is reversible and consequently can be compensated [120].

The laser tuneability, one of the major advantages of FEL sources, is obtained by merely modifying the magnetic field of the undulator in a given spectral range set by the electron beam energy. Operation at short wavelengths requires high beam energies for reaching the resonant wavelength, and thus long undulators (100 m range for 1 Å) and high electron beam density (small emittance and short bunches) for ensuring a sufficient gain.

The polarization depends on the undulator configuration. It can easily be changed from linear to circular, using APPLE-II or DELTA devices.

The different short wavelength FELs implemented around the world are shown in Fig. 6. In FELs, different configurations (oscillator, self-amplified spontaneous emission, seeding, echo) can be used.

In the oscillator mode [121] (Fig. 5a), the laser field, starting from synchrotron radiation, is stored in an optical cavity, enabling interaction with the optical wave on many passes. FEL oscillators cover a spectral range from the THz to the VUV, where mirrors are available [122]. The first FEL was achieved in 1977 on the MARK-III linac (Stanford, USA) in the infra-red in such a configuration [123]. ACO (Orsay, France) [124] provided the second worldwide FEL (first visible radiation) in 1983 and first FEL based harmonic generation [125].

Because of the limited performance of mirrors, short wavelength FEL are usually operated in the so-called self-amplified spontaneous emission (SASE) (Fig. 5b) setup [126], where the spontaneous emission at the input of the FEL amplifier is amplified, typically up to saturation in a single pass after a regime of exponential growth. Once the saturation is reached, the amplification process is replaced by a cyclic energy exchange between the electrons and the radiated field. The emission usually presents poor longitudinal coherence properties, with temporally and spectrally spiky emission, resulting from non-correlated trains of pulses, apart from the single spike operation, for low charge short bunch regime [127]. Thanks to recent accelerator advances (high peak current, small

Fig. 6. Map of FELs: in project in italic, in red for VUV soft X ray, in blue for hard X-ray. (For interpretation of the references to color in this text, the reader is referred to the web version of the article.)

energy spread, low emittance) and long undulator linac based single pass SASE FEL are blooming worldwide. They now provide tunable coherent sub-ps pulses in the UV/X-ray region, with record peak powers (MW to GW) and a substantial gain in peak and average brilliance. After LEULT (Argonne, USA) [128] in the VUV, FLASH (Germany) (30–4.5 nm) [14] operates for users, SCSS test accelerator (Japan, 40–60 nm) [15] is presently up-graded after serving users. In the Angstrom (Å) region, the first tunable fs X-ray FELs at 1.5 Å was achieved at the linear coherent light source (LCLS, Stanford, USA, 2 mJ, 14 GeV) [12] in 2009. LCLS employs one part of the existing SLAC room temperature linear accelerator at 14 GeV. Saturation has been achieved after 60 m of undulators. Recently, the energy has reached 6 mJ. Photon tuneability ranges between 1 keV and 10 keV. An out-of-vacuum DELTA undulator is under preparation for providing adjustable polarization. SACLA, the second worldwide X-ray FEL extending the radiation down to 0.06 nm, operates since June 2011 [13] (Japan, 8 GeV). SACLA operate with a thermo-ionic gun, a C band compact linear accelerator of 8 GeV, 18 5-m long adjustable gap in-vacuum undulators of 18 mm period length. Saturation starts after the 10th segment of undulator. Photon tuneability ranges between 5 and 20 keV, with energies up to 0.5 mJ. These X-ray FELs constitute the brightest X-ray beams ever produced on earth and have been already used by scientists. European XFEL [129] on a high repetition rate superconducting linear accelerator, the Korean XFEL [130] and the SwissFEL [131] are expected soon. Presently, no conventional laser can compete with the performance of LCLS or SACLA. These X-ray FELs, of a typical km length, use hundreds meters of undulators. Fifty years after the laser discovery, the emergence of several mJ X-ray lasers for users in the Angstrom range (the so-called fourth generation light sources) constitutes a major breakthrough, thanks to the accelerator and free electron lasers (FELs) developments and opens a new era for the investigation of matter, such as structure and function of biomolecules [2], electronic structure of atoms and molecules [3,132], non-equilibrium nuclear motion, disordered media and distorted crystal lattices [133,134], chemical reactions. Higher availability of X-ray pulses with stable energy, synchronized to an external pump laser, enabling jitter-free optical pump/resonant X-ray probe experiments will enable to step further.

For suppressing the spikes, improving the longitudinal coherence, reducing the intensity fluctuations and the jitter, a typical strategy consists in seeding (Fig. 5c) the FEL amplifier using an external seed that possesses the required coherence properties [135]. The seed can be an external laser wave or a short wavelength coherent light source, such as high order harmonics generated in Gas (HHG) [17], injected in order to interact with the electron beam in the undulator. Saturation is also more rapidly reached than in the SASE case, which makes the system more compact. In the high gain harmonic generation scheme (HGHG) [136] (Fig. 5d), an injected laser source induces the modulation in density of the electron bunch in the first undulator. The radiation is produced in the second undulator tuned on the harmonic of the injected wavelength. Coherent nonlinear harmonics of the fundamental wavelength are also generated. FEL pulse temporal and spectral distributions result from the seed itself and the FEL intrinsic dynamics. In particular cases, super-radiant modes exhibit further pulse duration narrowing and intensity increase [137]. Already in the early FEL times, an external laser source tuned on the undulator resonant wavelength was injected, enabling more efficient bunching and coherent harmonic generation with production of coherent radiation at 100 nm [138,139] in 1991. HHG seeding has been first performed on SCSS test accelerator at 160 nm [17] and at 60 nm [140], at SPARC with cascading demonstration [141] and at 30 nm at s-FLASH [142]. The only seeded FEL operated for users in the seeded configuration is FERMI@ELLETRA (Italy) [143], using a conventional laser as a seed. In this case as well, circular polarization is also provided to users, thanks to APPLE-II undulators.

Tuneability can be achieved on the injection source coupled to a gap change [143] or by applying a chirp (frequency drift) both on the seed and on the electron bunch [144]. Self-seeding can also be applied, in particular in the hard X-ray domain. A monochromator installed after the first undulator spectrally cleans the radiation before the last amplification in the final undulator [145]. Recently, self-seeding with the spectral cleaning [146] of the SASE radiation in a crystal monochromator appears to be very promising with the first results [147,148]. Frequency up-conversion can be very efficient, as demonstrated recently at FERMI@ELETTRA (from 266 nm to 4 nm) with a double cascade scheme [149].

In the echo enabled harmonic generation [150] (Fig. 5e) (EEHG) scheme), two successive laser–electron interactions are performed, using two undulators, in order to imprint a "sheet-like structure" in phase space. Higher order harmonics can be obtained in an efficient way. Echo has been experimentally demonstrated in the UV on the seventh harmonic [151] and up to the fourteenth [152] on the next linear collider test accelerator (SLAC) and on the Shanghai FEL test

facility [153]. It constitutes a breakthrough in up-frequency conversion from a conceptual point of view, and in terms of compactness and pulse properties (e.g. duration and wavelengths). Schemes derived from EEHG such as the triple mode chicane [154] open perspectives for very short wavelength (Å) and short duration at moderate cost.

Transverse modes result either from the resonator characteristics (in the oscillator configuration) [155], from the electron beam emittance (which should be of the order of the emitted wavelength), or from possible gain guiding on single pass FELs. Transverse coherence is usually deduced from double Young slits measurements and wavefront is measured with Hartmann sensors [156,157], providing a 3 nm wavefront residual on SCSS test accelerator at 60 nm.

Longitudinal coherence depends strongly on the configuration type. SASE regime leads to a spiky temporal and spectral distribution, with internal jitter and intensity fluctuations. It can somehow be improved by different means. Low charge short electron bunch operation can lead to single spike regime [158], but with a slightly reduced intensity. LCLS can operate in such a mode. An electron beam energy chirp (electron energy dependence along the bunch position) combined to undulator taper (variation of the peak field along the longitudinal direction) can also efficiently lead to a single spike FEL, as shown on SPARC [159]. Improved SASE, with a slippage enhancement by a chicane, can enable to control the longitudinal profile [160]. Besides, seeding is also quite efficient in reducing the spike and controlling somehow the laser longitudinal properties. The seed level should overcome the shot-noise [161] and this can become critical for a short wavelength seed. Seeding enables as well to get harmonics up to a higher order than in the SASE case [162]. Synchronization can indeed become critical [163]. Self-seeding with a single crystal monochromator [147] is efficient but particularly sensitive to energy fluctuations of the electron beam.

FEL harmonic generation efficiently shortens the delivered wavelength, using different schemes such as HGHG [135], fresh hunch technique where the light interacts in the second undulator in a non-heated part of the electron bunch [164], harmonic cascade [165], echo [150].

Two-color FEL, of interest for pump probe FELs, can be achieved by different means. First produced on CLIO in the infra-red in using two different undulator segments tuned at different wavelengths [166], it is now developed in single pass FELs. In the seeding case, one can fruitfully take advantage of the pulse splitting effect which can occur for particular seed pulse duration with respect to the electron bunch length [167], as shown on FERMI@ELETTRA with a chirped seed [168]. Thanks to the installed chicane for self-seeding and to the sufficient margin of undulator length, two colors are generated in tuning the two series of undulators at different wavelengths, the delay being adjusted by the chicane itself [169]. Further, a double slotted emittance spoiler enables to control the delay (fresh bunch) or in the iSASE configuration, undulators are slightly detuned to act as phase shifters. Direct X-ray splitting with a crystal is also provided in some cases.

Further FEL developments aim at providing radiation at even high photon energies, reaching higher intensities, with tapering [170,159] where the magnetic field of the undulator is adjusted to maintain the resonance condition with beam energy loss or the enhanced SASE [171] scheme where a first laser imprints a modulation in an undulator before being further accelerated in the next sections. FEL manipulation is foreseen with the different schemes (seeding, echo, single spike, modified SASE, XFEL oscillator...).

There is also a great interest in reducing the FEL pulse duration toward the attosecond range [172]. Various schemes have been proposed, such as emittance spoiler [173], the use of energy chirped electron beams used as a seed for a second stage [174] or with optical post-compression [175], selective amplification [176], electron energy modulation in a small part of the electron bunch with a few cycle laser [177], the combination of the slotted foil with eSASE [178], the generation of a few cycle XFEL [179]. In parallel to the quest of ultra-short pulses, very narrow spectral bandwidth (0.0001%) FELs are also of interest for particular applications. X FEL oscillators [180] on energy recovery linacs [181] can fulfill these requirements.

Multiple user operation is also a relevant challenge, for reducing the operating cost per experiment. Besides lowering the repetition rate by kicking consecutive bunches toward different FEL lines, superconducting linear accelerators, with the possibility to kick different parts of the electron bunch train to various FEL lines can provide a solution toward this objective, as proposed on EXFEL, NGLS [182] or LUNEX5 [183]. LUNEX5 (free electron Laser Using a New accelerator for the Exploitation of X-ray radiation of 5th generation) is a demonstrator project for investigating the production of short, intense, and coherent pulses in the soft X-ray region. LUNEX5 envisions a superconducting linac or a Laser Wake Field Accelerator, to be qualified in view of FEL application, a single FEL line composing the most advanced seeding configurations (HHG seeding, EEHG) and pilot user experiments to characterize and evaluate performance of these sources from a users' perspective for further optimization. Recently, it has been shown that the number of RF units effectively used for acceleration can be adjusted from bunch to bunch, and further kicked to different FEL lines [184]. The combined advantage of superconducting linear accelerators for high repetition rate and multiple FEL line requires however an adequate electron gun. The field of high repetition rate high brightness electron guns have recently progressed significantly [185], with in particular in the domain of the superconducting guns [186] already in use for the ELBE FEL.

It is also of interest to combine FELs with lasers and THz sources for pump-probe two-color user experiments.

5. Sources on novel accelerators

Besides improving the performances of FEL and the manipulation of their properties, another path of advance concerns the compactness. Besides complex seeding scheme and short period undulators, another path to explore is the change of accelerator type.

In laser wakefield accelerators (LWFA) [18], high-intensity laser pulses are focused in a dense plasma from a gas jet, gas cell or capillary discharge targets, and an ultrahigh longitudinal electric gradient is created. This ponderomotive force pushes the plasma electrons out of the laser beam path, separating them from the ions. A traveling longitudinal electric field, whose amplitude can reach several hundred GV/m is created, typically 10,000 times larger than conventional accelerators, the characteristic length scale of the wakefield, the plasma wavelength, being of 10–30 μm. In the so-called "bubble regime" [187–190], where a single laser pulse is used, electron beams in the 100 MeV range can be produced over mm distances, with energy spreads on the order of 5–10% and charge of hundreds of pC, or 1 GeV electron beam with somewhat lower charge with the laser pulse guided over a few cm in a capillary plasma discharge. Scaling laws predict that multi-GeV electron beams with nC charges might be attainable with the next laser generation [191] with an energy spread of a few %. Two-stage laser plasma accelerators have recently delivered GeV electron bunches with only 1% energy spread [192]. The colliding laser pulse [193] mechanism leads to 1–10% energy spread, 10–100 pC charges, 4 fs duration, with a stability range within 5–10% with control of the electron beam parameters such as charge and energy spread. The proposed cold injection technique could provide electron beam with energies from 0.3 to 1 GeV [194], few tenths of pC, a few fs

duration and a relative energy spread of less than 1% with a 200 TW laser.

This last decade, the reliability of LWFAs have been tremendously improved, delivering routinely electron beams [195] with typical current of a few kA [196], bunch length of a few fs, energy in the few hundreds of MeV to 1 GeV range [197], relative energy spread of the order of 1%, and normalized emittance of the order of π mm.mrad [198]. Such a LWFA appears as an attractive candidate for the next generation of colliders and for future compact light sources and FELs [21] with GeV electron beams, which provides an intermediate qualification goal before TeV LWFA colliders of interest in the long term for high energy physics. Using electron beams with the presently achieved performance in terms of energy spread and divergence however does not lead to direct FEL amplification whereas spontaneous emission from undulators has been observed [199]. Experiments are under way in various places (OASIS (Berkeley) [200], Strathclyde Univ. [201], MPQ [202], LOA/SOLEIL [203]...).

With respect to conventional accelerators, LWFA beams exhibit very different characteristics of phase space: in longitudinal, short bunch duration and large relative energy spread and in transverse, large divergence and micrometer size. The divergence can be handled by strong quadrupoles located very close to the electron source [204]. Electron beam manipulation by chicane decompression [205,206] or by the use of transverse gradient undulator [207] suggest that significant amplification with the present LWFA performance has become possible. It is for example one of the purposes of the LUNEX5 project, which is composed by besides the superconducting linear accelerator, a LWFA to be qualified with the FEL application in the quest toward ultra-short ultra-compact FEL sources. It could also enable straightforward operation of the FEL in the single spike regime for high coherence pulse delivery.

Dielectric accelerators [208] are also promising devices toward compact electron beam driver for FELs. A laser is injected into a dielectric structure, with typically 800 nm hole diameter where resonant spatial harmonic provides acceleration and non resonant spatial harmonics provides focusing. The galaxies [209] project intends to develop a FEL on such an acceleration.

Inverse free electron lasers [210], where an intense laser tuned to an undulator resonant wavelength, enables to raise the electron beam energy in a specific stage. Inversely to the FEL process, the laser gives energy to the electron beam. These inverse FELs can also serve to drive a short wavelength FEL.

6. Conclusion

HHG sources appear as mature, laboratory scale sources. They are evolving toward shorter wavelength, higher efficiency, higher repetition rate. They can provide pulses as short as attosecond duration. Storage ring lased synchrotron light sources are mature, they evolve toward full transverse coherence with ultimate storage rings and tailored bunches. They still present rather long bunches and intermediate energy spreads. Energy recovery linacs are presently in the phase of test facility development, they are very promising for providing intense ultra-short X-ray user facilities. With respect to synchrotron radiation, free electron lasers provide further longitudinal coherence, by setting emitters in phase. In particular, linac based SASE sources are the first coherent tunable light sources for users in the hard X-ray range, with an intermediate longitudinal coherence since trains of emission can be uncorrelated. Seeded soft X-ray free electrons lasers, coupled to harmonic generation, are also successfully operating for users as well and enables to provide full longitudinal coherence in the VUV and soft X-ray emission. FELs tend now toward high photon availability sources, evolution toward advanced "tailored" characteristics with muti-color, adjustable polarization, higher powers and energies... High order harmonics generated in gas and on solid targets, synchrotron radiation, free electron lasers are complementary sources, offering a large flexibility on light source properties. Clearly, further synergy and interplay between accelerator based light sources and lasers will provide new possibilities and flexibilities. Indeed, common concepts such as seeding have already been identified. New acceleration schemes and related technologies are also emerging, even though demonstration experiments, improvement of stability and reliability is still required.

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