The advent
of X-ray free electron lasers

Free Electron Lasers (FEL) use free electrons in the periodic permanent magnetic field of an undulator as a gain medium. They extend from far infrared to X-rays, they are easily tunable and provide a high peak power. The advent of tunable intense (few mJ) short pulse (down to the attosecond regime) FELs with record multi GW peak power in the X-ray domain enables to explore new scientific areas. These unprecedent X-ray sources come along with versatile performance.

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Vacuum tubes in which free relativistic electrons interact with electromagnetic waves were rapidly developed last century. First observed in particle accelerators in 1947, the synchrotron radiation emitted by accelerated charged particles, in bending magnets for example, is collimated in a narrow angular cone ($1/\gamma$ with γ the Lorentz factor) and spectrally expands from far infra-red to X-rays. High intensity is achieved with alternated magnetic poles, as in undulators, that create a periodic permanent magnetic field: The radiation from the different periods N_u can constructively interfere and emit on-axis a spectrum of sharp lines at the resonant wavelength and its n^{th} order odd harmonics $\lambda_n = \lambda_u(1+K_u^2/2)/2n\gamma^2$, with λ_u the undulator period, B_u peak magnetic field, K_u the deflexion parameter $= 0.94\ \lambda_u\ (\text{cm})\ B_u(\text{T})$. First observed in 1951, undulator radiation is well collimated, adjustable in polarisation and is tunable by a change of the electron energy or by the undulator magnetic field. A. Einstein first discussed the stimulated emission from excited atoms in black-body studies in 1917. The MASER achieved in 1954 by C. Townes, replaced the electron beam amplification of vacuum tubes by stimulated emission of excited molecules introduced in a microwave cavity resonant at the molecule transition frequency. A. Schawlow and C. Townes proposed to use a Fabry-Perot type resonant cavity to extend the radiation to the optical spectral range [1], giving birth to "optical lasers", later named LASER (Light Amplification by Stimulated Emission of Radiation) and underlined limits in wavelength reduction for which new approaches are needed.

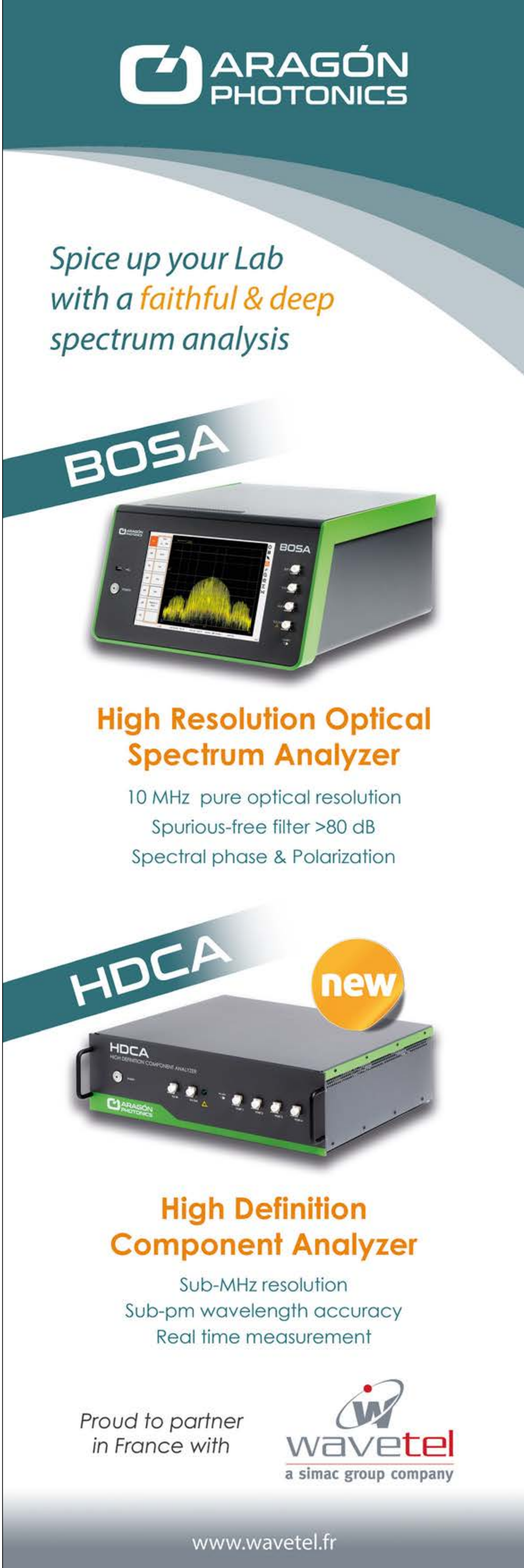
J.M.J. Madey from Stanford University searched for a Free Electron Radiation mechanism that could be coupled to the optical cavity approach of the lasers. He opted for Stimulated Compton Scattering process and replaced the counter-propagating photon beam by a virtual one with the radiation from high charge electrons in a strong periodic transverse magnetic field, for being more efficient. The scheme (see Fig.1) includes the electron beam in the undulator field as the gain medium and the laser type optical resonator. After Madey's first gain calculations in quantum mechanics, a classical theory was found to be applicable in most cases. The undulator synchrotron radiation, stored in the optical resonator, exchanges energy with the electrons, leading to an electron energy modulation that is gradually transformed into microbunching at λ_n separation. Electrons set in phase thus radiate coherently, the light is then amplified to the detriment of the electron kinetic energy. The gain of the medium is proportional to the electronic density, the cube of the undulator length and the inverse of the cube of the electron beam energy. FEL wavelength is merely tuned by the electron beam energy of the magnetic field of the undulator. Short wavelength operations require high electron beam energies according to the undulator resonance condition, and thus require high electron beam performance and high undulator lengths. Saturation takes place by enhancement of energy spread, or by unsatisfied resonance condition due to electron beam energy decrease to the benefice of the optical wave. The undulator length is also limited by the slippage of the radiated photons travelling slightly quicker than the electrons, for not the radiation to escape from the electron bunch.

Low gain FELs

J. M. J. Madey pursued at Stanford University on a superconducting linear accelerator with FEL experimental demonstration in 1976 of the amplifier regime and in 1977 of the oscillator one at 3.4µm [2]. Six years later, FEL oscillators were produced on the ACO (France) storage ring in the visible, in the infra-red again at Stanford and at Los Alamos (USA) linac (9-11µm). Coherent harmonic generation (see Fig.1), with a seed laser tuned on the undulator resonant wavelength extended to the UV and VUV. Linac based FEL output power was enhanced by undulator tapering (i.e. adjusting the magnetic field value along the longitudinal coordinate), in maintaining the FEL resonance condition with the electron beam energy decrease. The FEL oscillators, also installed on Van de Graafs, microtrons, energy recovery linacs, were largely developed, and cover from infra-red to VUV spectral range (190nm on the ELETTRA storage ring FEL). They offer a very high degree of coherence (the transverse one thanks to the optical resonator and the longitudinal approaching the Fourier limit thanks to multi-passes). The short wavelength operation is limited by the gain value compared to the mirror losses submitted to drastic irradiation conditions.

High gain FEL development

Along with high gain FEL studies, the production of coherent radiation from a self-instability, in the Self Amplified Spontaneous Emission (SASE) regime without optical cavity was discovered (see Fig.1) [3]. The FEL starts from the undulator spontaneous emission shot noise : electrons communicate together through the radiation and the space charge field and "self bunch" on the scale of the radiation wavelength periods. They emit collectively coherent synchrotron radiation in areas with nearly equal phases driving for a collective instability. After a "lethargy" period required for the initial pulse to build up, the light is amplified exponentially with a gain length L_g depending on the



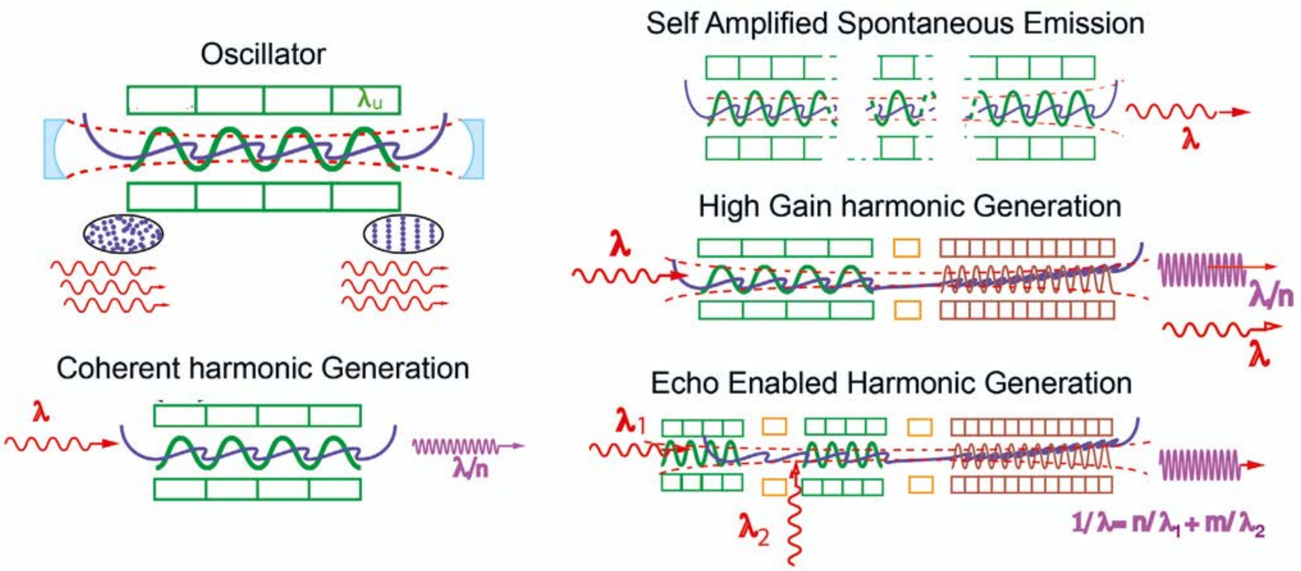


Figure 1: FEL gain medium (relativistic electrons (trajectory in blue) in the undulator (in green)) for the different configurations: oscillator (spontaneous emission stored in an optical cavity, with a sketch of the energy modulation and electron bunching), harmonic generation with an external laser (red) tuned on the undulator resonant wavelength, SASE (undulator spontaneous emission amplified in a single pass), High Gain Harmonic Generation, Echo Enable Harmonic Generation with two electron / laser interactions.

Pierce parameter ρ_{FEL}, that also characterises the FEL efficiency and gain bandwidth. Saturation is typically reached after $20L_g$. The interaction between the electrons is only effective over a cooperation length (slippage in one gain length). The uncorrelated trains of radiation lead to spiky longitudinal and temporal distributions and poor longitudinal coherence, apart from single spike operation.

SASE was first experimentally demonstrated in the mid-eighties in the far infrared where the gain is more favourable in view of the electron beam quality available at that time and then in the infrared one decade later. Thanks to the accelerator developments for future colliders, a major step was crossed with the use of a photo-injector with improved electron beam properties with respect to thermionic guns, on the Los Alamos experiment, demonstrating five orders of magnitude amplification and saturation at 12µm. The beginning of the 21st century saw the advent of the saturated SASE in the visible and UV on the Low-Energy Undulator Test Line (LEUTL, Argonne National Laboratory, USA) (530 and 385nm) in 2000, VISA SASE FEL (USA) (423 - 281nm) in the VUV on FLASH (Germany) (109nm) with a low emittance high charge photo-injector in 2001, *i.e.* 25 years after the FEL invention. Tunability in the 80-120nm range was demonstrated, with a very high degree of photon beam transverse coherence. With higher peak current, the GW level (~1µJ) had been reached in the 95-105nm spectral range [4]. These SASE results competed the shortest wavelength achieved on a storage ring FEL oscillator, making a turning point in

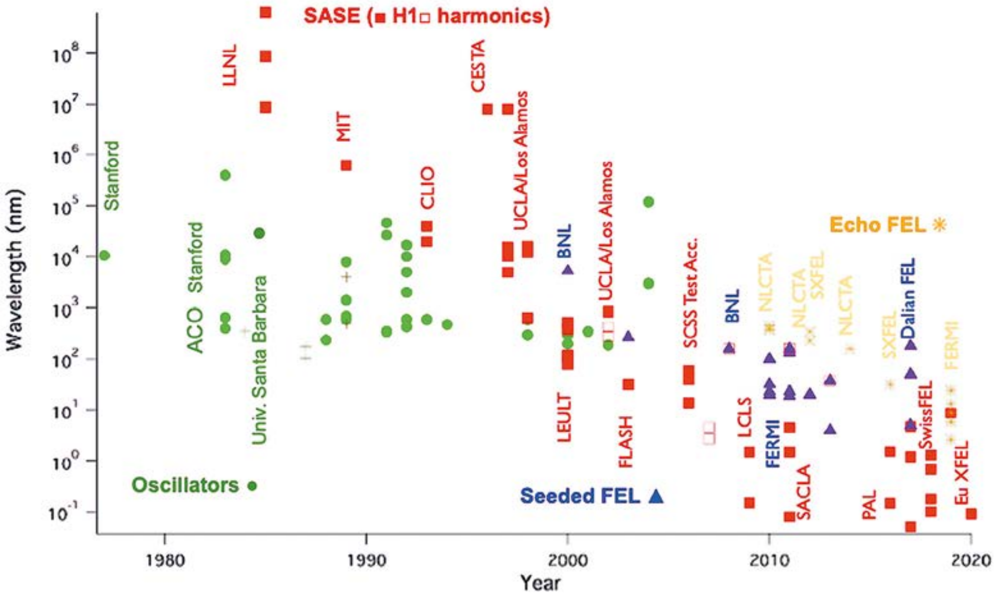


Figure 2: FEL evolution versus years: achieved FEL wavelengths versus year for various configurations (oscillators (non exhaustive), coherent harmonic generation, SASE, seeding).

the choice of the FEL accelerator driver (from storage rings to linear accelerators) and configuration (from oscillator to single pass) for short wavelength FELs. 2000 appeared to bring a transition where VUV was reached both by oscillator and SASE configurations (see Fig.2).

The XFEL advent

The path towards the X-ray domain with SASE radiations was paved with new achievements in the soft X-ray region on the SCSS Test Accelerator (Japan) (60-40nm, 30mJ), FLASH (6.5, 4.1 and water window), which was established as a user facility in 2005. Then, the advent of hard X-ray FELs opened a new area, one decade later, with LCLS in Stanford (USA) at 0.15nm, with saturation after 60m of undulators [5] in 2009, more than forty years after the first FEL in the infra-red in Stanford, SACLA (Japan) in 2011 down to 0.08nm, PAL FEL (Korea) in 2016, Swiss FEL (Switzerland) and European XFEL (Germany) at high repetition rate in 2017 [6] (see Fig.3). European XFEL, an international facility, is driven by a 17.5GeV superconducting linear accelerator with up to 5000 electron bunches per second that serves different FEL branches and experimental stations, providing 2mJ pulses and 6W average power. High repetition rate XFELs enabling multi-users operation are also under preparation on LCLSII (USA) and SHINE (China) projects. In order to alleviate for SASE pulse jitter, spiky spectral and temporal distributions that are prejudicial for use, several configurations are employed: Single spike mode with low-charge short electron bunch regime, chirped electron bunch associated with an undulator taper, seeding with an external laser spectrally tuned on the undulator fundamental radiation, that enables to reduce intensity fluctuations, saturation length and improve the longitudinal coherence (see Fig.1) [7]. Direct seeding has been extended to short wavelengths with High order Harmonics generated in gas (HHG). Nonlinear harmonics can also be applied for efficient up-frequency conversion with cascades of "modulator"/ "radiator" undulators ("modulator" insuring electron modulation with the seed, "radiator" emitting FEL on fundamental and harmonics wavelength), the first cascade using the external seed, the second using the harmonic emitted by the previous radiator with a conversion order up to ~15th order per stage. FERMI@ ELLETRA (Italy), the first seeded FEL user facility with two FEL branches: FEL1 (100 - 20nm single cascade) and FEL2 (20-4nm, double cascade harmonic generation), has reached an up-frequency conversion by 192. The seeded Dalian FEL (China) covers the 50-150nm range. For the X-ray domain where seeds are not available, seeding is performed with the FEL itself: a monochromator installed after a first undulator series spectrally cleans the radiation before the last amplification in final undulators. The Echo Enabled Harmonic Generation scheme that imprints a sheet-like structure in phase space via two successive electron-laser interactions in

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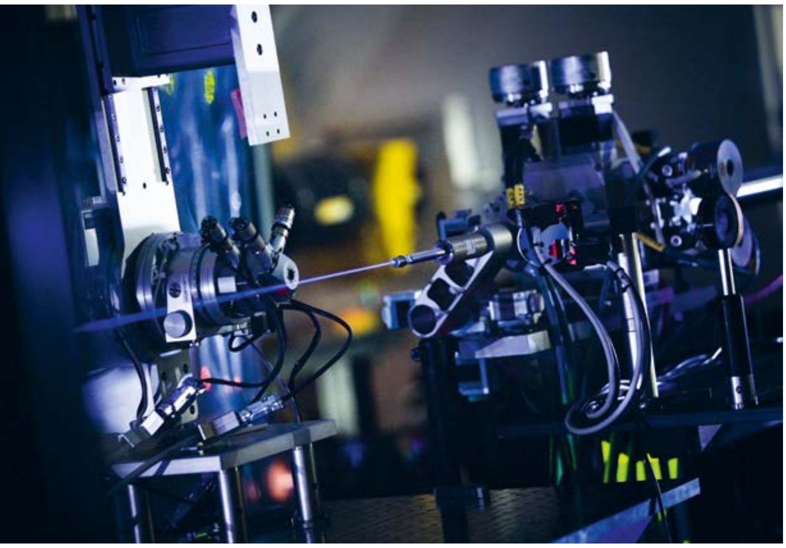

Figure 3: Picture of the European X FEL.
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two undulators has demonstrated efficient up-frequency conversion. Coherent emission down to harmonic 101 and lasing down to harmonic 45 at FERMI [8], with enhanced stability as compared to HGHG configuration, multi-colour operation (5.7 and 5.9nm), radiation could be extended to the water window. Hard X-ray FELs employ high beam energies for reaching the resonant wavelength, long undulators (0.1-1km) and high electron beam density (small emittance and short bunches) for ensuring a sufficient gain. These intense XFELs largely benefited from the improvements of the electron beam performance, thanks to the development of photo-injectors and future colliders.

User applications of XFELs

Since the laser invention, the FEL advent in the X-ray domain half a century later opened new areas for matter investigation (structure and dynamics) on unexplored domains with higher temporal resolution [9]. Ultra-intense XFELs give access to the unexplored domain of X-ray nonlinear optics under extreme conditions. In addition, the femtosecond XFEL can be combined to an optical laser for pump (manipulating the internal electronic state)/probe experiments, enabling to provide molecular movies (tracking of structure and electronic states) and process dynamics. Besides, taking advantage of the coherence and of the femtosecond FEL duration, and considering that the diffraction can take place before the destruction of the sample, coherent diffraction imaging can be applied to tiny, fragile crystals in solution even at a high repetition rates (serial crystallography) and single particles such as virus with very good spatial resolution below 1nm. XFELs permits the imaging of living cells and the dynamics of proteins (for example, conformation change of the chromophore in the photoactive yellow protein) can be followed by pump-probe measurements.

Versatile performance of the XFELs facility

Various advanced manipulations enable to XFEL properties to be adapted to the user needs. XFELs provide ultra-short single spike SASE pulse using various electron beam manipulation shaping or FEL specific regime. Single 280 (480) attosecond pulses at 0.9 (0.5) keV [10] with a peak power exceeding 100GW have been recently achieved and used. These features, unlikely to be reached by HHG in a near future, open the path to unique exploration of electron dynamics with X-ray nonlinear spectroscopy and single-particle imaging [16]. After the early two-colour FEL oscillators, XFELs are also operated with two different pulses delayed in time and spectrally shifted for pump-probe experiments, using various schemes (use of one single bunch and differently tuned undulators, pulse splitting combined with chirp or twin bunches). Polarization is controlled on demand and optical vortices can also be produced. FEL oscillators come back into play for the X-ray regime for high repetition rate low bandwidth XFELs or for driving kW average power EUV lithography.

Prospects with new accelerator concepts

FEL is also adequate to qualify new alternative accelerator concepts (dielectric acceleration, inverse FEL, plasma acceleration) for which electron beam performance still does not meet those achieved by conventional accelerators. An amplification of two orders of magnitude at 27nm was recently achieved on the laser plasma accelerator at SIOM (China) [11].

Conclusion

The advent of X-ray Free Electron Laser implemented on conventional linear accelerators, 40 years after the FEL invention constitutes a second laser revolution, enabling to decipher unexplored area of structure and dynamics of matter and biological samples. The unprecedent combined XFEL performance, combining single attosecond pulse, multicolour, GW power and high repetition rate make them unique tools in the landscape of X-rays light sources. ●

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