The quest for brilliance: light sources from the third to the fourth generation

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It is evident that light is perhaps the most important tool by which we know the world around us. This is true not only for our everyday experience, but also for the scientific pursuit of an understanding of nature. Experiments using electromagnetic waves, in a range of wavelengths going well beyond the relatively small range of visible light, have played an important role in the development of modern science. Atomic and molecular spectroscopy, i.e. the study of the characteristic wavelengths emitted by matter in the gas phase, has been fundamental in establishing the laws of quantum mechanics and has given us important information on the composition of stars and planets. Einstein's brilliant 1905 interpretation of the photoelectric effect has opened the way to photoemission spectroscopy, which is still today one of the most important probes of the electronic structure of solids and solid surfaces, using UV light. Röntgen's discovery of x-rays in 1895, and the subsequent 1911 demonstration of x-ray diffraction by crystals, by von Laue and others, laid the foundations of crystallography, by which we can unravel the atomic structure of crystals. Every secondary school student is familiar with Watson and Crick's exploit of 1953 which identified the double helix structure of DNA from the x-ray diffraction work of Franklin and Wilkins, probably the most famous piece of crystallographic work ever.

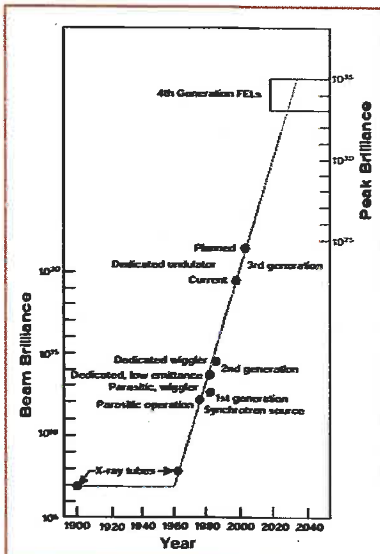
It is therefore hardly surprising that scientists have been eager to obtain the brightest light sources, in order to understand the atomic and electronic structure of matter better and better. In parts of the infrared, in the visible and in the near UV, the invention of the laser has provided an extraordinary tool, which has led to many exciting discoveries and applications. Over a much broader range, encompassing not only the infrared and the visible, but also the whole of the UV and the x-rays to wavelengths well below 0.05 *nm*, the tool of choice for many scientists is synchrotron radiation (or *synchrotron light*) i.e. the bright emission of highly collimated electromagnetic waves from electrons (or positrons) orbiting at ultrarelativistic energies in storage rings with diameters of tens to several hundreds of meters (generally called with the slightly improper term *synchrotrons*). In spite of their large dimensions and associated cost, there are some 50 or so storage rings around the world built and operated solely for the purpose of producing light, and more are under construction.

Although synchrotron light is ever so popular today, the basic phenomena underlying this way of producing light are by no means very recent or novel. The electromagnetic theory of Maxwell, which very naturally contains aspects later formalised in the special theory of relativity, is all that is needed to predict the basics of synchrotron light. The radiation of accelerated charges, e.g of charged particles following a circular trajectory at constant speed, and subject therefore to centripetal acceleration, has a very different angular distribution depending on whether the speed of the particles is very small compared to the speed of light c (the classical limit), or very close to c (the ultrarelativistic case). In the latter situation, electromagnetic theory shows that the light emitted by the particles is confined to a narrow cone. The angular opening of the cone is given by $1/\gamma$, where $\gamma = E/mc^2$ is the ratio of the energy of the orbiting particle compared to its rest energy. Spectral analysis of the emitted radiation further shows that the emitted frequencies comprise all harmonics of the revolution frequency of the particle around the ring, from the fundamental up to harmonics of order $\sim \gamma^3$, above which the intensity drops rapidly. For an electron or positron of 1 GeV energy, γ is almost 2 000, whence the sub-*mrad* collimation of synchrotron light, and its broad spectrum, typical revolution frequency around large rings being in the *MHz* range, and $\gamma^3 \sim 10^{10}$–10^{12} for electron energies of a few GeV. In addition to these remarkable properties, synchrotron light also has well defined polarisation properties. To understand why the light is so bright one has to consider how large the acceleration is for a particle which is going at essentially the speed of light in one direction and that half a circumference(i.e. typically ~ 1 μs) later is moving at the same speed in the *opposite* direction!

The first description of the emisssion of relativistic particles following a circular orbit is given in 1912 by Schott, and received little attention. The problem became of high interest in the 1940's, when it became apparent that the main hurdle in the construction of higher and higher electron energy accelerators is the rapid growth of the radiated power with electron energy (proportional to E^4 for a fixed radius of the machine). The modern derivation of the basic expressions used in the description of synchrotron radiation was provided by Schwinger in the USA and by Ivanenko and Pomeranchuk in the Soviet Union. The first direct observation of synchrotron radiation was reported in 1947 by Elder *et al.* at the 70 MeV Synchrotron in the General Electric Laboratories in Schenectady, NY (USA) [1].

First, second and third generation light sources

The driving force behind the development of light sources is the optimization of their *brilliance* (or spectral brightness), which is the figure of merit of many experiments. Brilliance is defined as a function of frequency given by the number of photons emitted by the source in unit time in a unit solid angle, per unit surface of the source, and in a unit bandwidth of frequencies around the given one. The units in which it is usually expressed are *photons/s/mm²/mrad²/0.1%BW*, where *0.1%BW* denotes a band-



◂ **Fig. 1:** (after D. Moncton) Historical evolution of the brilliance of 8 keV x-ray sources, extrapolated into the future. The left scale represents the average brilliance (averaged over the pulsed structure of the synchrotron beams), the right scale refers to the peak brilliance (at the top of one pulse).

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▲ **Fig. 2:** View of Elettra, a third generation light source in Trieste.

width $10^{-3}\omega$ centered around the frequency ω. As one can appreciate from the definition, brilliance puts a premium not only on the photon flux (photons per second in a given bandwidth), but also on the high phase space density of the photons, i.e. on being radiated out of a small area and with high directional collimation. Liouville's theorem ensures that brilliance is a property of the source and not of the optics of the beamline which delivers the photons to the experimental station. An ideal set of optical elements can only preserve the brilliance, a real one will always degrade brilliance (as some photons get lost on the way down the beamline). The brilliance of available synchrotron sources has been growing at a formidable pace in the last decades (see Fig. 1), since the first attempts at a systematic exploitation of storage rings as sources of photons for scientific experiments in the 1960's.

At that time, some electron storage rings designed and built for nuclear and subnuclear physics started to be used *parasitically*, for some fraction of the time, as sources of photons for experiments in atomic, molecular and solid state physics. These machines are nowadays referred to as "first generation light sources". The experimental results were so interesting and promising to stimulate the construction of dedicated rings, designed and optimised to serve exclusively as light sources. Examples of these "second generation" machines are the BESSY I ring in Berlin, the two National Synchrotron Light Source rings in Brookhaven, NY (USA), the SuperACO ring in Orsay, near Paris, and the Photon Factory in Tsukuba (Japan). In the 1990's a new generation of rings started operation. These "third generation" synchrotron sources are characterized, as we shall briefly explain below, by a reduced emittance (i.e. reduced phase space volume) of the circulating particle beam, and by the extensive use of undulators as radiation sources, with a further increase of the brilliance by several orders of magnitude. Examples of this generation of sources are the European Synchrotron Radiation Facility (ESRF) in Grenoble, the Advanced Light Source in Berkeley, California, Elettra in Trieste (see Fig. 2), BESSY II in Berlin, Max-II in Lund, Sweden, the Advanced Photon Source, in Argonne, Illinois, Spring 8 in Japan, and the Swiss Light Source in Villigen (CH).

Two basic advances, as has been mentioned before, made the transition to the third generation possible. On the one hand, the progress in the design and fabrication of the lattice of magnets that guide the electrons (or positrons) around the storage ring allowed a reduction in the size of the cross section of the circulating particle beam, and a better collimation (parallelism of the velocities of all particles at a given point in the ring). This can be expressed quantitatively as the volume of the phase space projection along the horizontal plane of the particles' trajectory (horizontal emittance). This quantity is measured in *nm rad* (nanometers times radians). It is typically of order 30-100 *nm rad* or more in a second generation machine, and typically of order 5 *nm rad* in a third generation ring. It is intuitive that the properties of small beam cross section and high collimation of electrons translate into corresponding properties of the radiated photons and therefore contribute to the brilliance. The second advance is the development of insertion devices (wigglers, and most importantly undulators) which are arrays of magnets, typically 2 to 5 *m* long, inserted in a straight section of the ring, and which produce a vertical magnetic field with a sinusoidal dependence along the electron trajectory (see Fig. 3). The resulting Lorenz force on the drifting electrons modifies their straight trajectory into a zig-zag one, producing a large number of bends with intense radiation emission. Wigglers are large field devices, which produce deviations of the beam from the straight trajectory that are large when compared to the characteristic angle of synchrotron radiation emission, i.e. $1/\gamma$. The radiation cones from each bend of the trajectory do not overlap and the wiggler multiplies the number of bends, preserving the same broad spectrum of emitted wavelengths. The undulator, on the other hand, is a small field device, where the largest deviation of the electrons from the straight line is at most of order $1/\gamma$, so that radiation cones from the whole trajectory of an electron spatially overlap, and the resulting electromagnetic fields interfere. Detailed calculations show that the result of the interference is to enhance emission at a particular wavelength and at its harmonics and to suppress it elsewhere. These wavelengths are given by:

$$n\lambda = (\lambda_0 / 2\gamma^2)(1 + K^2/2),$$

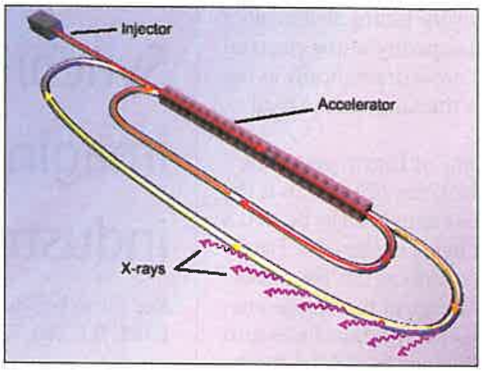
where λ_0 is the period of the sinusoidal magnetic field of the device, $K = \gamma\,\theta$, θ being the electrons maximum angle of deviation from the straight trajectory, and $n = 1, 2, 3, \ldots$ is the order of the harmonic.

Interference phenomena in undulators, therefore, concentrate the brilliance around a discrete set of wavelengths. It is however very important to notice that the fundamental wavelength is tunable by the mechanical movement of the magnet arrays: when their distance varies, so does the maximum magnetic field and therefore the maximum deviation angle θ and the K parameter. Undulator radiation can be characterized as quasi-monochromatic and *tunable*.

There are about ten years of experience in the operation of third generation light sources. Examples of the breakthroughs which were made possible by these remarkable scientific instruments in a variety of fields, encompass condensed matter physics, materials science, chemistry, mineralogy, biology and even medical and industrial applications. Certainly more will follow in the years to come; nonetheless scientists have been wondering about the next step, i.e. the concepts necessary to bring about a further increase in brilliance by a few orders of magnitude.

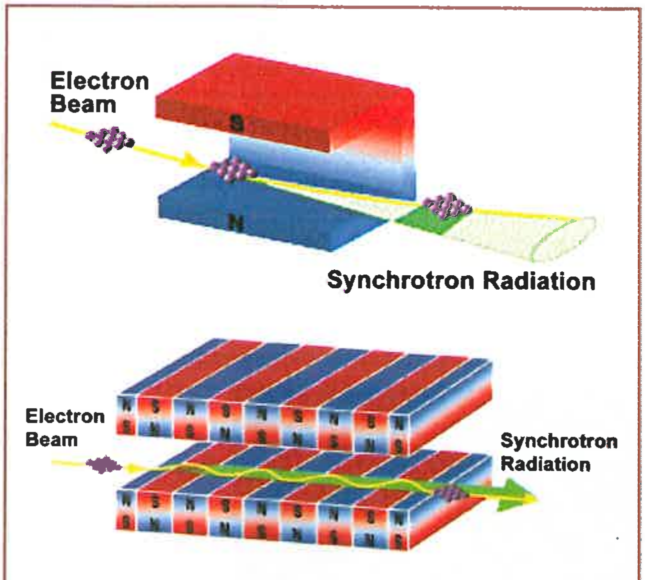
Fourth generation light sources: Free Electron Lasers and recirculating machines

One feature of the storage ring as a light source is that the same electrons turn for hours and hours, going hundreds of thousands and even millions of time per second through the same undulators and dipole magnets. The radiofrequency cavities in the ring give back the radiated energy to the electrons (they typically radiate about 0.1% of their energy at each turn) so that they keep turning and turning. Every times an electron emits a photon, a non-deterministic quantum process, recoil effects perturb its momentum and position. These millions and millions of "random" perturbations per second determine a lower limit to the emittance that the magnetic lattice of the ring can impose on the electrons, i.e. they prevent the phase space volume of the electrons from being too small. A careful and thorough quantitative analysis show that it is not possible to lower substantially the emittance of a storage ring below the values achieved in third generation machines. This is the reason why the pursuit for the fourth generation has oriented itself to "single-pass" or "few-passes" machines, where a given electron goes only very few times through an undulator.



◀ **Fig. 4:** Scheme of principle of an energy-recovery Linac. Electrons from the injector are accelerated in the Linac, then recirculated in the Linac a first time via the first arc section to be accelerated to the final energy; at their maximum energy, electrons circulate in the larger arc section where they radiate into many beamlines, before returning to the Linac to give back most of their energy to the RF cavities.

One possibility is to abandon the ring geometry and consider a linear accelerator, or Linac, which feeds the accelerated electrons into a long undulator. At the end of a Linac, it is foreseeable - and to some extent already experimentally proven—that very low emittance and also very short electron bunches (~ 100 fs, as opposed to the ~ 50 ps typical for storage rings) can be produced. The generation of correspondingly short x-ray photon pulses was recently demonstrated in Stanford, in the SPPS project at the SLAC Linac, where bright ~ 80 fs x-ray pulses were generated by a standard undulator [2]. But a carefully designed and very long undulator at the end of a suitable Linac can produce radiation with full spatial coherence, i.e. laser-like and delivered in sub-ps pulses of extremely high peak brilliance (exceeding 10^{29} - 10^{30} *photons/s/mm²/mrad²/0.1%BW*, to be compared with ~ 10^{21} for third generation undulators). This is the extraordinary promise of Linac-based Free Electron Lasers (FEL's). The difference between "spontaneous" or "incoherent" undulator radiation and FEL radiation is that in an undulator, as was briefly explained, there is constructive interference between the fields radiated by one electron at different points of its trajectory. In the FEL process, the fields of *different* electrons also interfere constructively. This is possible if the distribution of electrons within the bunch is not uncorrelated, but there are density modulations with the wavelength of the undulator resonance. If the undulator is long enough and the bunch has suitable properties, these density fluctuations can be imprinted by the radiation itself. Consider indeed the progress of a bunch of electrons down the undulator. The light emitted at the back of the bunch goes a little bit faster than the electrons (the latter travel at a speed slightly lower than c, and have a wiggly trajectory due to the magnetic field, while photons go straight!). The light wave, therefore travels down the bunch and, interacting with the electrons, it imparts a density modulation at its wavelength. In point of fact, the process is helped by the spontaneous density fluctuations statistically arising inside the electron bunch: the travelling light wave enhances dramatically those of the corresponding wavelength. The modulated bunch radiation starts to grow exponentially down the undulator length, until saturation is reached. This mechanism is known as SASE, or Self-Amplified Spontaneous Emission and produces full spatial coherence for radiation of very short wavelength. This has been demonstrated at DESY in Hamburg for wavelengths down to 80 *nm* [3], using the superconducting Linac technology, and progress towards the goal of 6 *nm* is underway.



▲ **Fig. 3:** (Top) Schematic view of the generation of synchrotron radiation in a dipole bending magnet, where electron bunches are deviated to follow the orbit around the ring.
(Bottom) Generation of synchrotron radiation in an insertion device, where the upper and lower magnet arrays produce vertical fields forcing the electrons to follow a zig-zag trajectory.

The idea of pushing this project to 0.1 *nm* within a European X-ray FEL Laboratory, on the model of the ESRF, is presently being discussed. A similar project (the LCLS, "Linac Coherent Light Source") using the room-temperature SLAC technology is underway in Stanford.

There are also many smaller projects to generate VUV and soft x-ray coherent radiation, also based on linear accelerators, each of them with some specific feature that makes them quite complementary to one another. Some of these do not envisage reliance on statistical fluctuations to induce the initial density modulation in the electron bunch, but aim to induce it externally, with a pulsed laser synchronised to the electron bunches. It turns out in fact that an electron bunch in the magnetic field of the undulator behaves as a strongly non-linear optical medium. An external laser of wavelength λ can generate density modulations with components not only at λ but also at the harmonics λ/n, so that a visible or near UV conventional laser can generate modulations of much shorter wavelengths. The light of a first undulator could induce even shorter wavelength modulations in a second one, in a harmonic cascade process. The advantage of these "seeded" FEL schemes is the very precise control of the pulse timing, as the FEL pulses are triggered by the predictable pulses of the external laser, and not by the random noise of the density fluctuations in the electron bunch. An example of such a project is the FERMI@ELETTRA seeded FEL, with the ultimate goal of achieving 10 *nm*.

The scientific scenarios opened by 100 *fs* long, spatially coherent pulses of UV and x-radiation are very exciting. This explains the large number of projects that are being pursued or proposed at various laboratories around the world. Clearly the technical difficulties connected with the requirements on the quality of the electron bunches (emittance, peak current, etc.) grow dramatically as the required wavelength decreases towards the range of the hard x-rays.

In the pursuit of the higher brilliance of fourth generation sources there is an alternative road to the Linac FEL's . This is the approach based on recirculating Linacs, as exemplified by the 4GLS ("4th Generation Light Source") project being designed at Daresbury Laboratory in the UK. The idea is to combine the "few passes" strategy to lower emittance with the advantage of the ring geometry in which a large number of tangential beam lines and experimental stations can be exploited by users in parallel. It consists of a Linac that feeds electrons into one or several arc-shaped sections on which undulators, wigglers and even FEL's can be inserted (Fig. 4). At the end of the (last) arc, electrons are either dumped or fed back into the Linac, where they can return part of their energy to the RF fields in the accelerating cavities (this latter version is called "energy-recovery Linac"). The Linac is preferably of the superconducting type, one of the reasons being that it can support a much higher repetition rate (a much smaller time interval between successive bunches).

Conclusions

The interest of scientists from a variety of disciplines in ever more brilliant light sources in the UV and x-ray spectral region is not decreasing. While the third generation sources are still delivering a wealth of interesting science, the intense activity to generate spatially coherent, ultrashort pulses promises a new frontier to be opened a few years from now. It will be extremely interesting to monitor the progress of the many projects underway all over the world.

About the author

Massimo Altarelli was trained at the University of Rome as a condensed matter theorist. At the ESRF in Grenoble, he was Research Director (1987-1993), and Head of the Theory Group (1994-1998). Since 1999 he has been in Trieste on the staff of the International Centre for Theoretical Physics, and Director of the Elettra Synchrotron Light Source.

References

[1] An excellent account of the early history of synchrotron radiation, with many references, can be found on pages 27-35 of E.-E. Koch, D.E. Eastman and Y. Farge, "Synchrotron radiation – a powerful tool in science", in *Handbook of Synchrotron Radiation*, vol 1A, E.-E. Koch editor (North Holland, Amsterdam, 1983) pp 1-63;

and also at the site http://xdb.lbl.gov/Section2/Sec_2-2.html, which provides an account by A.L. Robinson, as part of the X-ray Data Booklet Project of Lawrence Berkeley National Laboratory.

[2] See http://www-ssrl.slac.stanford.edu/jhhome.html for a short description of the state of the project

[3] J. Rossbach, Nucl. Instr. Meth A475, 13 (2001); for a first experimental application, see H. Wabnitz *et al.*, Nature 420, 482(2002)

Synchrotron radiation imaging and diffraction for industrial applications

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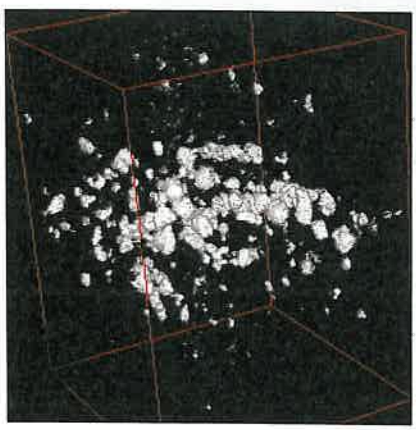
The use of the modern synchrotron radiation (SR) sources for X-ray imaging and diffraction provides new possibilities for applied science and technology. They include an enhanced resolution (spatial and/or temporal), the possible use of the coherence of the beam, in-situ experiments and accurate quantitative measurements of local density or strain. Selected applications showing the improved capabilities in absorption or phase microtomography, time-resolved diffraction experiments showing the way a lithium-based battery works, or strain measurements on stir-welding materials, are presented as examples. The combination of diffraction and imaging (i.e. diffraction with micron spatial resolution) allows further applications, such as the study of grain growth, or the X-ray topographic investigation of defects responsible for spurious modes in single crystal resonators.

The aim of the present paper is to highlight the contribution of synchrotron radiation X-ray imaging and diffraction to the investigation of industrial topics. These techniques, which allow the visualization of the volume of systems opaque for other probes and the determination of their structure, have been dramatically renewed by the use of modern, "third generation", synchrotron radiation (SR) facilities beams. Real time/high-resolution experiments are performed for both X-ray imaging and diffraction. The high energy of the available beams allows the deformation within bulk materials to be investigated. The high coherence of the beam allows phase contrast imaging to be exploited, the contrast arising from phase variations across the transmitted beam. Absorption and phase microtomography provide three-dimensional information on features like cracks, porosities or inclusions, which are of high interest for applied topics such as metallurgy, polymers, or reservoir rocks containing fuel. The spatial resolution of these techniques is now in the 1 μm range. Lastly the combination of diffraction and imaging, through the new tracking techniques, or the use of Bragg diffraction imaging (X-ray topography) provides additional information on industrially interesting materials.



◄ **Fig. 1:** 3D image of the distribution of holes (in white) within a copper target (transparent), 269*284*201 pixels, 2 μm voxel size