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Synchrotron radiation: third generation sources

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Abstract
The advent of synchrotron radiation has expanded and revolutionized the use of x-rays as a research tool in a way which could not have been foreseen 40 years ago. In this contribution, we concentrate on the essential elements of third generation synchrotron sources. Their outstanding properties have convinced researchers in 'small' science that it is worth the effort to leave their home laboratories and to perform some of the most demanding, as well as rewarding research at a big centre. The many orders of magnitude gained in brilliance over rotating anode x-ray tubes make all the difference.

1. Introduction

When Röntgen discovered x-rays just before the beginning of the 20th century, it took him and others only a few months to realize their eminent importance for the investigation of medical and applied problems [1]. In the first 50 years after this event, the intensity of these x-ray tubes was increased by about two orders of magnitude until the classical method of producing x-rays reached its natural technological limits with the rotating anode tube. This was sufficient for most medical applications, for radiography with structural resolution down to 0.1 mm and crystallography on crystals with a reasonable size. The application of x-rays became so widespread that e.g. more than 50 000 participants come together every year at the annual meeting of the American Radiological Society.

In x-ray tubes, x-rays are emitted by decelerating electrons in e.g. a copper anode. The emission cone spans the full half space. With the exception of radiology on large objects, only a small fraction of this emission cone is used; the rest is thrown away. If samples are small and irradiation occurs with good directional resolution, as e.g. in crystallography, these sources are far from ideal (see figure 1). The figure of merit for such application is rather brilliance than flux. Brilliance is determined by the number of x-ray quanta per area of the source, per solid angle, per second and often per spectral interval.

In the 1950s, synchrotrons and later storage rings for electrons came into operation which emit x-rays due to a large centripetal acceleration. At relativistic energies, this acceleration can become enormous while, unlike in an anti-cathode, the electrons are not lost. In addition, the relativistic transformation from the reference frame of the emitter, the electron, to the

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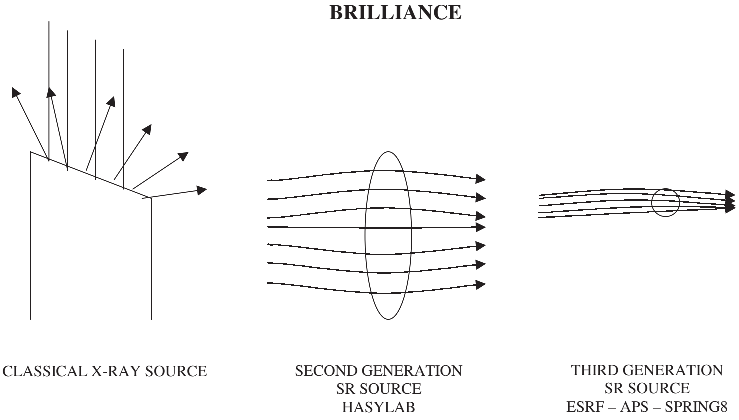


Figure 1. Increase of brilliance due to better collimation and smaller source size in going from a classical x-ray source to second and third generation synchrotron radiation sources.

Table 1. The main parameters of the three third generation synchrotron radiation sources.

	ESRF	APS	SPRING8
Nominal energy (GeV)	6	7	8
Circumference (m)	844	1104	1436
Nominal beam current (mA)	200	100	100
Lifetime (hours)	60	29	50
Horizontal emittance ε_H (nm rad)	3.8	7.6	7.0
Vertical emittance ε_V (pm rad)	30	100	7 (?)
First users/BL commissioning	1994	1996	1998

laboratory system squeezes the emission pattern into a narrow cone. This evidently favours the brilliance of such sources of synchrotron radiation (see figure 1).

Until about 1975, synchrotron radiation was used practically exclusively as a by-product of machines designed and built for elementary particle physics. During these first years, the pioneering experiments in many different research fields demonstrated the usefulness of this new tool. A community of users formed worldwide which realized that they could do even better if the machines were optimized and built from scratch as x-ray sources. The idea of third generation sources was born (second generation sources were those machines which had been built for particle physics but were handed over to the synchrotron radiation community). The increase in brilliance over the century as given in figure 2 clearly demonstrates the importance of synchrotron radiation.

At this moment, three third generation storage rings are in operation worldwide (see table 1): the European Synchrotron Radiation Source (ESRF) in Grenoble, France, the Advanced Photon Source (APS) in Argonne, USA, and SPring-8 in Harima, Japan. This contribution will mainly highlight the special properties of these third generation x-ray sources taking the ESRF as an example.

2. Optimization of the machine parameters

The goal of a so called dedicated synchrotron radiation source was clearly set by the definition of brilliance. It meant minimizing the cross section and the divergence of the electron beam

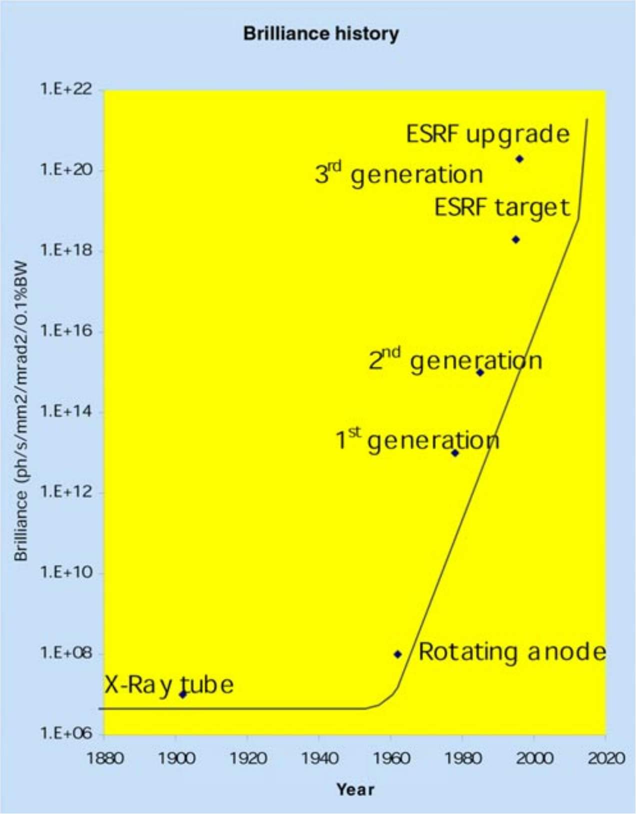


Figure 2. Evolution of brilliance of x-ray sources during the 20th century.

which is the source and impregnates its characteristics on the x-ray beam. A further measure is to increase the current and optimize the acceleration pattern. The latter means an optimization of magnet structures and a carefully considered choice of the beam energy. All these parameters are not independent from each other. Quite simply, the increase of the current has its limitations since electrons are confined in phase space and filling too many electrons into a given phase space volume will blow up the beam. In addition, a too high electron density shortens the lifetime of the stored beam due to electron electron scattering.

It would surpass the goal of this contribution to explain here in detail all the considerations which had to be taken into account to minimize the emittance of the electron beam which can be considered for the purpose of this article as the product of size times divergence. There are excellent reviews available [2].

A few items may nevertheless be highlighted:

(i) The bigger the circumference and the lower the energy, the smaller the emittance.

(ii) Vibrational and drift stability of the magnet structure becomes an essential element for the functioning of these machines. The beam diameter can be as small as 25 μm FWHM, and beam motion must remain less than a fraction of this.

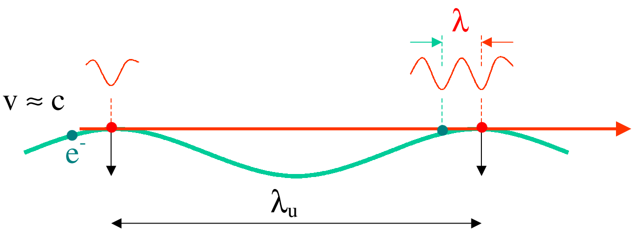
(iii) If all elements (bending magnets, quadrupoles, sixtupoles, . . .) are carefully aligned, such a machine, however, can function in some respect with fewer problems than a machine with a large beam. The field errors close to the axis of e.g. a quadrupole magnet are negligible.

One of the most important achievements of machine designing was, however, the provision of dispersionless straight sections along the circumference which serve as the location of so-called insertion devices (wigglers, undulators, wavelength shifters). It took some theoretical effort and even more practical experience to find out that it is possible to design and operate storage rings in such a way that a large number of these locations of 5 m length or more can be accommodated and that insertion devices can be switched on and off almost without influencing the performance of the machine at other locations. From this achievement, a concept of modern synchrotron radiation source can be derived where the storage ring is considered as the 'power plant' and the straight sections are the 'outlets' into which insertion devices can be plugged in as 'appliances'.

3. Wigglers and undulators

One of the most decisive innovations which made the high performance of third generation sources possible was the introduction of wigglers and undulators [2]. The idea is fundamentally simple. A magnet lattice which changes its direction many times forces the electron beam on a sinusoidal trajectory. There is a common tangent to all these wiggles such that the emission intensity is multiplied by the number of wiggles. This can give rise to a factor of 100. Even better, for certain wavelengths, under certain conditions, the amplitudes superimpose coherently and 100 wiggles can produce an increase of 10 000 in the spectral brilliance. In reality, the factor is rather of the order of 2500 due to the broadening of the undulator peaks through the energy spread of the stored electrons.

UNDULATOR EFFECT



$$\lambda = \frac{1}{n}\,\frac{\lambda_u}{2\gamma^2}\,(1+\tfrac{1}{2}K^2)$$

$$K \approx \lambda_u[\mathrm{cm}]B_0[\mathrm{T}]$$

Figure 3. The principle of the undulator effect due to the superposition of wave trains emitted at successive wiggles of an undulator.

The most interesting type of insertion device is now definitively the undulator. An electron moving on a sinusoidal trajectory with a typical period length of 20 mm emits electromagnetic radiation with a phase depending on the direction of the curvature. The emissions from all the bends have a well defined phase with respect to each other, and may superimpose for specific wavelengths λ. There is a perfect analogy to the treatment of an optical grating. At first sight, one is astonished that a macroscopic structure with 20 mm period length displays interference at the 1 Å level. It is, however, easily explained as pointed out in figure 3 that the difference in advance of the phase of the wave to the emitting electron at successive undulator periods is the distance which matters. This distance is small due to the fact that electrons move almost with the velocity of light. If electrons as light moved on a straight line, the 1 in brackets in the equation describes this contribution. The second contribution in the bracket is due to the detour the electron takes by moving on a curved line. K depends on the magnetic field and thus opens the possibility to tune the undulator peaks. One should note that even with third generation machines, the maximum amplitude of the undulator trajectory remains a fraction of the beam size.

Undulators are now built in a large variety. The most common way is to install permanent magnet structures on both sides of the beam. By reduction of the gap, the magnetic field is increased. Gaps are limited to about 10 mm by the vacuum chamber. If this is too large (shorter period lengths), the magnet structure has to be placed into the vacuum. Then the gap can be further closed (to 5 mm at the ESRF). The limitation is given when the magnet structure starts to scrape the electron beam. Since some of the flexibility which ESRF has acquired by subdividing the 5 m long straight sections into three segments of 1.6 m each is lost, ESRF keeps the number of in-vacuum undulators low. With fixed narrow vacuum chambers, different types of undulator may be installed and exchanged within hours. On the other hand, the smaller gaps can be very useful for high energy photon emission and high intensities (see figure 4). SPring-8 uses in-vacuum undulators on a large scale.

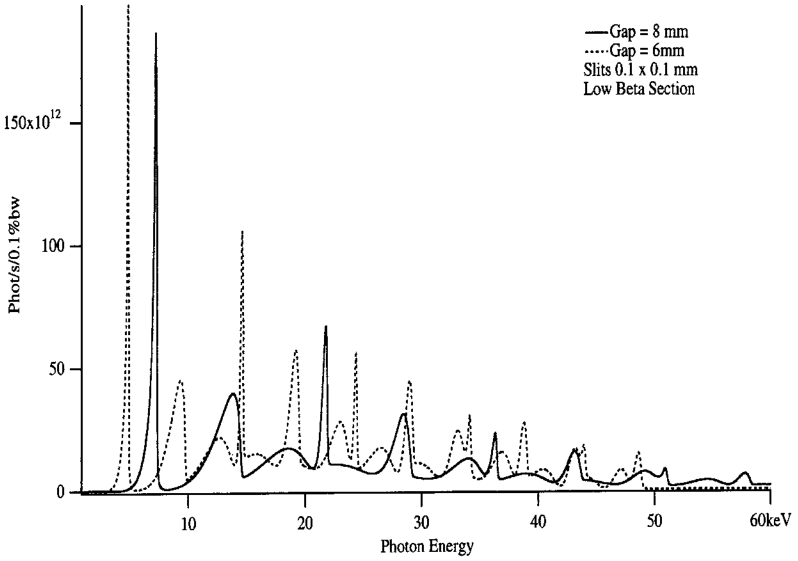


Figure 4. The on-axis spectrum of a powerful in vacuum undulator. The sharp peaks are the uneven harmonics 1, 3, 5, 7, ... while the even harmonics 2, 4, 6 are also observed due to the finite acceptance of the monochromator of ≈0.02 mrad. (Thanks to P Elleaume, ESRF.)

In spectroscopic experiments, the undulator gap and thus the peak in the spectrum is varied synchronously with the energy selection of the monochromator. This technique has been brought to maturity at specific beamlines and allows us to obtain optimum conditions also for this type of experiment. The higher harmonics from an undulator were originally not produced with sufficient efficiency. A technique of 'shimming' was developed at the ESRF to make the undulator field perfectly periodic. Figure 5 shows the gain in intensity of the higher harmonics. Note that only the even harmonics show up when the beam is observed in the forward direction. This follows from the Fourier transform of the 'unit cell', the sinus.

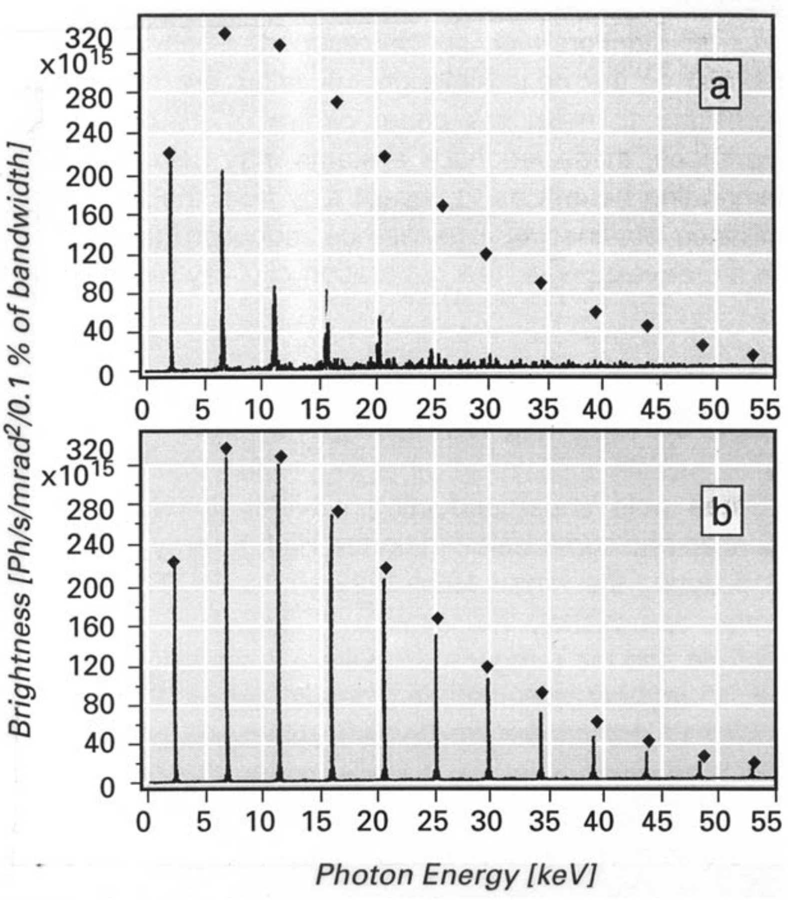


Figure 5. Comparison of undulator spectra before (a) and after (b) spectrum shimming. Spectrum shimming increases the brightness at all harmonics and removes background between peaks. The dots correspond to an ideal field. (Thanks to P Elleaume, ESRF.)

It is interesting to note that an undulator emits this clean spectrum only in the forward direction in a cone whose half width is given by $\sqrt{\lambda/L}$ where L is the length of the undulator. Any emission outside this cone is shifted to longer wavelengths (as can be worked out using the optical grating analogy) and falls off in intensity. The typical use of undulator radiation is in any case restricted to an angular range of about 10 μrad in order not to increase the apparent source size. Note that one needs to look 'head on' at the up to 5 m long source. Nevertheless, there is also emission away from the forward direction which may amount to several kW and needs to be cooled away (figure 6).

If magnetic fields become very high and the K value in the equation in figure 3 approaches 10, the undulator effect is still occurring, but the first harmonics have moved into the soft x-

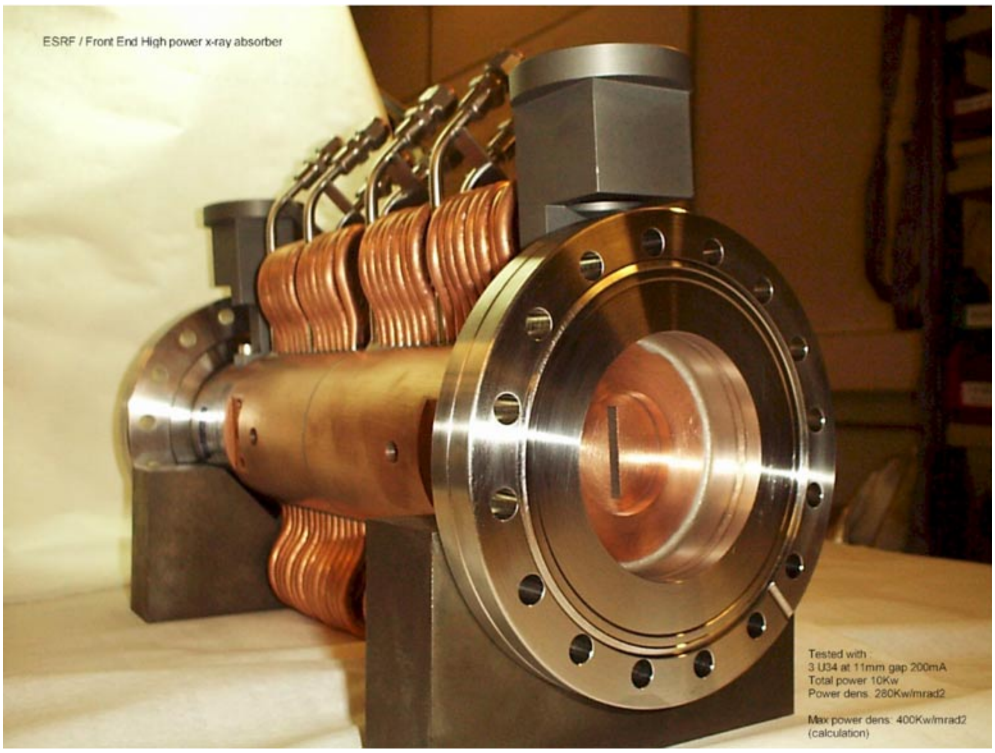


Figure 6. The high power front-end absorber developed at ESRF, which allows us to cool away up to 18 kW of unwanted power, which is emitted off-axis.

ray range. Then the main intensity is in the very high harmonics which merge into an x-ray continuum. The device is then called a wiggler (for the x-ray range).

4. Assets of the new sources

4.1. Scattering from small samples

Everything mentioned above about brilliance immediately highlights this possibility. Since diffraction from crystals after the discovery of von Laue, Friedrich and Knipping in 1912 became the outstanding method to obtain the atomic arrangement in inorganic and organic molecules, any new substance needs to be crystallized. Being able to obtain results from smaller and smaller crystals saves enormously on such efforts and makes structure analysis possible in many cases for the first time. Synchrotron radiation offers the additional advantage of providing, with the multiwavelength anomalous diffraction (MAD) technique, a means to overcome the phase problem. High intensity in a small beam additionally promotes high pressure research in diamond anvil cells, stress strain analysis in single grains of technical materials etc.

4.2. Spectroscopy

The tunability of synchrotron radiation over the full energy range of the x-ray edges of all stable elements allows for a variety of atom specific spectroscopies. The chemical binding of an element is typically reflected in small but detectable shifts of the edges and in characteristic structures, near the edges (XANES). The distances to the nearest neighbors are reflected in the extended x-ray absorption fine structure (EXAFS), which may span an energy range of several hundred eV above the edges. It is the interference structure in the wave function of the emitted and partially backscattered photoelectron.

4.3. Resonant scattering

This technique combines scattering with spectroscopy. As was mentioned already with the specific MAD technique of protein crystallography, certain atoms in a structure can be manipulated to change their scattering power by choosing specific wavelengths at their edges. It has been demonstrated that it is also possible to enhance the contribution of specific orbitals with which these atoms bind and thus obtain the spatial arrangement of electronic orbitals. While the interpretation is not simple, such techniques look extremely promising.

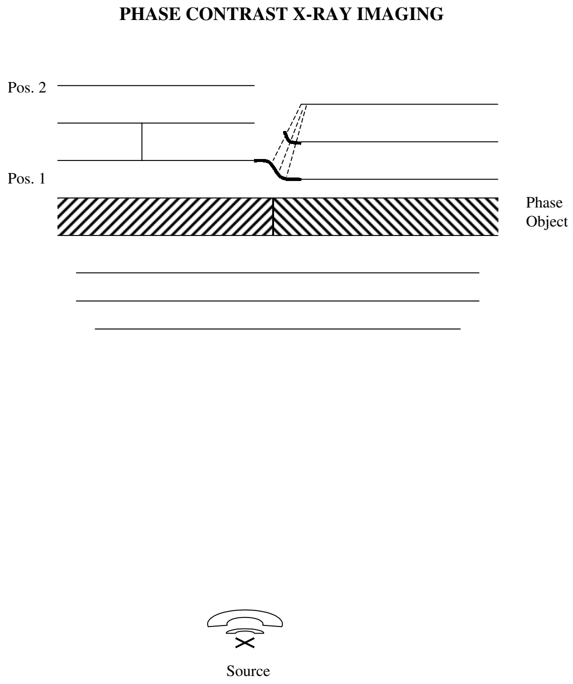


Figure 7. Phase contrast imaging. How to see the boundary between two materials of different index of refraction in an object due to the distortion of the wave front.

4.4. Circular and linear polarization

Undulator radiation as it is emitted in the plane of the undulation is linearly polarized. Specific undulators which force the electrons on a spiraling path produce right or left handed circular polarized x-rays. Their interaction with matter is specifically sensitive to the magnetic structure. The same holds if the change in the direction of polarization can be analysed after the scattering process. In combination with resonance scattering, this has become another powerful technique to elucidate complex magnetic structures. But circular radiation can also exhibit

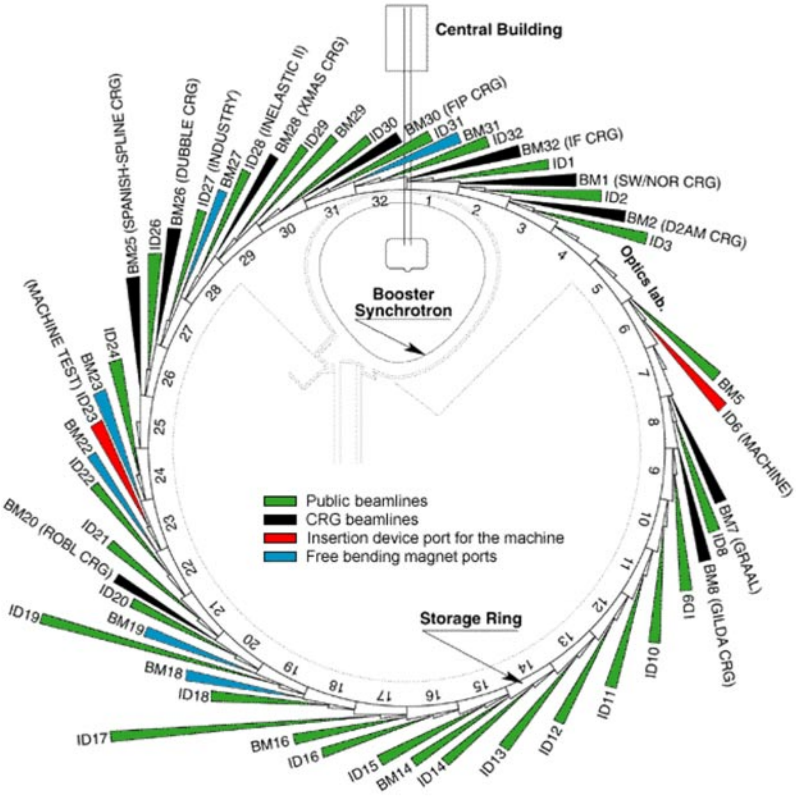


Figure 8. The arrangement of beamlines around the ESRF storage ring. There are 32 equivalent unit cells, each allowing in principle for one insertion device and one bending magnet beamline. Out of the 32 insertion device locations, five are blocked for acceleration, injection and diagnostics. (From *Highlights ESRF 1999*.)

dichroism which is a technique to investigate non-magnetic naturally occurring handedness in molecular crystals.

4.5. Inelastic scattering

The high brilliance of the source allows us to use high resolution backscattering monochromators with resolution in the 1 meV range at 20 keV photon energy. This opened up the field of inelastic scattering of x-rays with well defined momentum transfer similar to that of neutron scattering. Neutrons are still better in resolution at low energy transfer but their kinematic range is limited and problems can now be tackled in a complementary fashion. Dynamical properties of non-crystalline materials, liquids and glasses are the main target.

4.6. Coherence

Longitudinal coherence is determined by the length of a wave train which depends on the monochromacy which one imposes. Using the nuclear resonance scattering from ^{57}Fe, a coherence length of 30 m can be obtained. With an ordinary monochromaticity of 10^{-4}, it is much less.

Table 2. List of ESRF public beamlines in operation.

Beamline	Dedication
ID1	Anomalous scattering
ID2	Soft condensed matter
ID3	Surface diffraction
ID9	Time dependent, high pressure
ID10A	Troika U universal, speckle correlation
ID10B	Troika II, membranes
ID11	Materials science
ID12A	Circular dichroism
ID12B/ID8	Polarization dep. photoemission
ID13	Microfocus, soft condensed matter
ID14A	Protein crystallography 1, EH1
	Protein crystallography 1, EH2
ID14B	Protein crystallography 2, EH3
	Protein crystallography 2, EH4
ID15A	High energy diffraction
ID15B	High energy inelastic scattering
ID16	Inelastic scattering I
ID17	Medical
ID18	Nuclear scattering
ID19	Topography, coherent imaging
ID20	Magnetic scattering
ID21	X-ray microscopy
ID22	Microfluorescence, imaging
ID24	Dispersive EXAFS
ID26	X-ray absorption on ultra-dilute samples
ID27	Industry, TXRF
ID28	Inelastic scattering II
ID30	High pressure
ID32	Surface analysis
BM5	Optics, universal
BM14/ID29	Multiwavelength anomalous diffraction
BM16/ID31	Powder diffraction
BM20	X-ray absorption spectroscopy

Transverse coherence ℓ depends on the source size s and the distance D from the source D as $\ell = (\lambda/s)D$. This results in $\ell \approx 50\ \mu$m at the ESRF. It is the lateral distance of structures in a sample which can scatter coherently in such a way that the interference of the two waves is detectable.

As an example, any classical x-ray tube at reasonable distance has enough transverse coherence to resolve the Laue spots. This only requires a lateral coherence of 100 lattice constants. With a third generation synchrotron source, the coherent diffraction from colloidal suspensions of fairly large particles in solution is resolved. Furthermore, their motion gives rise to phase changes and thus to intensity fluctuations. This initiated the field of x-ray speckle spectroscopy.

Moreover, lateral coherence can be interpreted in another way as the region over which the wavefront emitted from the source is sharp and undistorted. Any non-absorbing object which exhibits variations in the index of refraction will distort this wave. The structures become visible according to the scheme of figure 7 at some distance from the object. This opens up a new type of imaging supplementing the classical amplitude contrast of radiography by

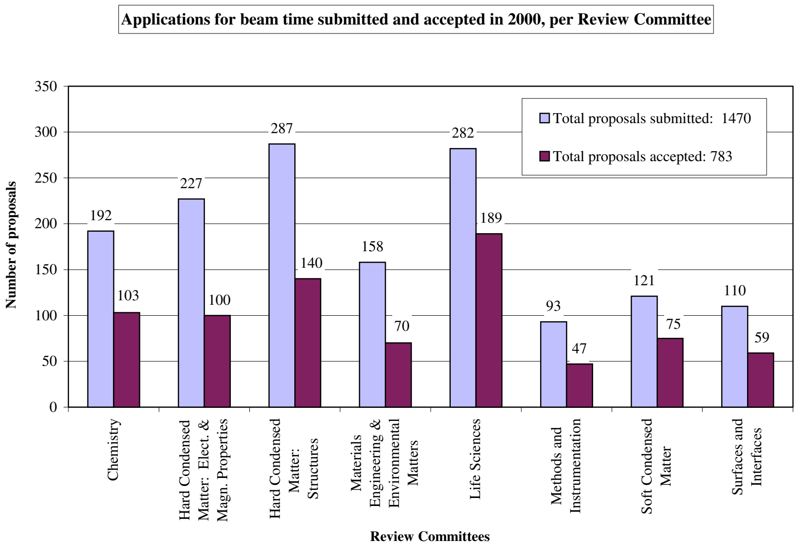


Figure 9. The number of proposals received and allocated in the year 2000 sorted according to different scientific areas.

phase contrast. Phase contrast occurs in objects which are thin and consist of light atoms. This specifically holds for organic tissue which cannot be differentiated in ordinary medical radiography.

5. User operation

There can obviously be no doubt that third generation synchrotron radiation sources are extremely useful for fundamental and applied research. The machines are fairly large and complex installations, and each of them has to serve a scientifically and geographically widespread community. Indeed, as mentioned above, there are presently only three installations in operation world-wide.

The mode of operation can be chosen differently. APS has chosen to rent out whole sections of the machine to a consortium of users who usually then equip and operate it for a variety of applications under their responsibility. ESRF restricts this mode of operation to about ten bending magnet beamlines which are built and operated by the CRGs. All the insertion device beamlines are managed centrally by the institution and each of them is dedicated either to a method or to a scientific field. This allows an optimization in each case and is considered to have been very successful. Figure 8 shows the arrangement of the beamlines, and table 2 indicates their dedication. Six years only after its opening, the institute is in full operation. New ideas can only be realized by rededicating existing beamlines.

The distribution of users over the different scientific fields is shown in figure 9. The applications go to the eight review committees which recommend the acceptance of the best proposals.

6. Conclusion

A full picture of the importance of third generation sources would require a scientific review of the different fields and the way they have benefited from the first six years of operation of such machines. This goes far beyond the scope of this introduction. No mention is made here of the whole field of beamline optics and specifically how it is possible to handle collimated x-ray beams with a primary power of up to 1 kilowatt without destroying the high brilliance. This would have needed to go into the details of monochromator cooling, crystal and mirror optics. For specific applications, some other contributions to this volume will give some examples.

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