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8. SYNCHROTRON CRYSTALLOGRAPHY

8.1. Synchrotron-radiation instrumentation, methods and scientific utilization

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8.1.1. Introduction

Synchrotron radiation (SR) has had a profound impact on the field of protein crystallography. The properties of high brilliance and tunability have enabled higher-resolution structure determinations, multiple-wavelength anomalous-dispersion (MAD) techniques, studies of much larger molecular weight structures, the use of small crystals and dynamical time-resolved structural studies. The use of SR required development of suitable X-ray beamline optics for focusing and monochromatization of the beam, which had to be stable in position and spectral character, for rotating-crystal data collection. Finely focused polychromatic beams have been used for ultra-fast data collection with the most advanced SR sources, where a single bunch pulse of X-rays can be strong enough to yield Laue diffraction data. The optimal recording of the diffraction patterns has necessitated the development of improved area detectors, along with associated data-acquisition hardware and data-processing algorithms. Sample cooling and freezing have reduced and greatly diminished radiation damage, respectively. In turn, even smaller crystals have been used. The low emittance of SR sources, with their small source size and beam divergence, corresponds well with the small size and low mosaicity of protein crystal samples. The evolution of SR source brilliance each year over the last twenty years has changed by many orders of magnitude, a remarkable trend in technical capability.

8.1.2. The physics of SR

The physics of the SR source spectral emission was predicted by Iwanenko & Pomeranchuk (1944) and Blewett (1946), and was fully described by Schwinger (1949). It is 'universal' to all machines of this type, *i.e.*, wherever charged particles such as electrons (or positrons) travel in a curved orbit under the influence of a magnetic field, and are therefore subject to centripetal acceleration. At a speed very near the speed of light, the relativistic particle emission is concentrated into a tight, forward radiation cone angle. There is a continuum of Doppler-shifted frequencies from the orbital frequency up to a cutoff. The radiation is also essentially plane-polarized in the orbit plane. However, in high-energy physics machines, the beam used in target or colliding-beam experiments would be somewhat unstable; thus, while pioneering experiments ensued through the 1970s, a considerable appetite was stimulated for machines dedicated to SR with stable source position, for fine focusing onto small samples such as crystals, and with a long beam lifetime for more challenging data collection. Crystallography has been both an instigator and major beneficiary of these developments through the 1970s and 1980s onwards. The evolution of new machines and the massive increase in source brilliance, year after year, are shown in Fig. 8.1.2.1(*a*); the most recent additions are SPring-8 (8 GeV) and MAX2 (1.5 GeV), thus illustrating the need for a range of machine energies today (Fig. 8.1.2.1*b*). A general view of an SR source as exemplified by the SRS at Daresbury is shown in Fig. 8.1.2.2. An example of a machine lattice (the ESRF) is shown in Fig. 8.1.2.3.

The properties of synchrotron radiation can be described in terms of the well defined quantities of high flux (a large number of photons), high brightness (also well collimated), high brilliance (also a small source size and well collimated), tunable, polarized, defined time structure (fine time resolution) and exactly calculable spectra. The more precise definitions of these quantities are

$$\text{Flux} = \text{photons per s per } 0.1\%\ \delta\lambda/\lambda, \tag{8.1.2.1a}$$

$$\text{Brightness} = \text{photons per s per } 0.1\%\ \delta\lambda/\lambda \text{ per mrad}^2, \tag{8.1.2.1b}$$

$$\text{Brilliance} = \text{photons per s per } 0.1\%\ \delta\lambda/\lambda \text{ per mrad}^2 \text{ per mm}^2. \tag{8.1.2.1c}$$

Care needs to be exercised to check precisely the definition in use. The mrad^2 term refers to the radiation solid angle delivered from the source, and the mm^2 term to the source cross-sectional area.

Another useful term is the machine emittance, ε. This is an invariant for a given machine lattice and electron/positron machine energy. It is the product of the divergence angle, σ', and the source size, σ:

$$\varepsilon = \sigma\sigma'. \tag{8.1.2.2}$$

The horizontal and vertical emittances need to be considered separately.

The total radiated power, Q (kW), is expressed in terms of the machine energy, E (GeV), the radius of curvature of the orbiting electron/positron beam, ρ (m), and the circulating current, I (A), as

$$Q = 88.47E^4 I/\rho. \tag{8.1.2.3}$$

The opening half-angle of the synchrotron radiation is $1/\gamma$ and is determined by the electron rest energy, mc^2, and the machine energy, E:

$$\gamma^{-1} = mc^2/E. \tag{8.1.2.4}$$

The basic spectral distribution is characterized by the universal curve of synchrotron radiation, which is the number of photons per s per A per GeV per horizontal opening in mrad per 1% $\delta\lambda/\lambda$ integrated over the vertical opening angle, plotted versus λ/λ_c. Here the critical wavelength, λ_c (Å), is given by

$$\lambda_c = 5.59\rho/E^3, \tag{8.1.2.5}$$

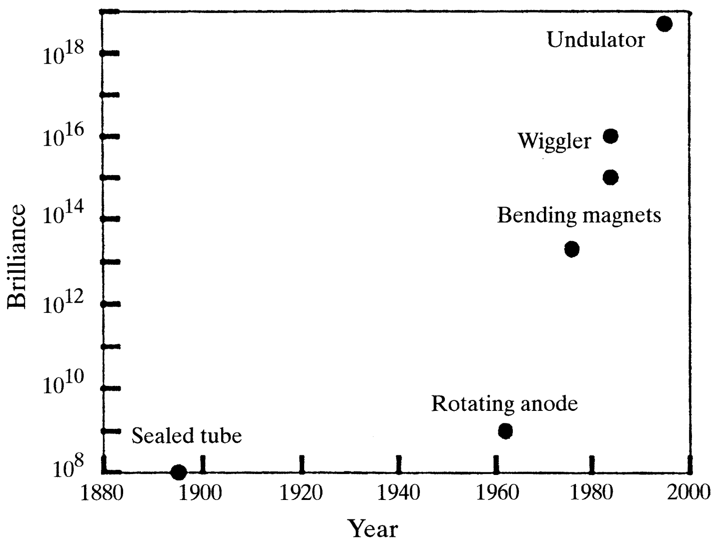
again with ρ in m and E in GeV. Examples of SR spectral curves are shown in Fig. 8.1.2.4(*a*). The peak photon flux occurs close to λ_c, the useful flux extends to about $\lambda_c/10$, and exactly half of the total power radiated is above the characteristic wavelength and half is below this value.

In the plane of the orbit, the beam is essentially 100% plane polarized. This is what one would expect if the electron orbit was visualized edge-on. Away from the plane of the orbit there is a significant (several per cent) perpendicular component of polarization.

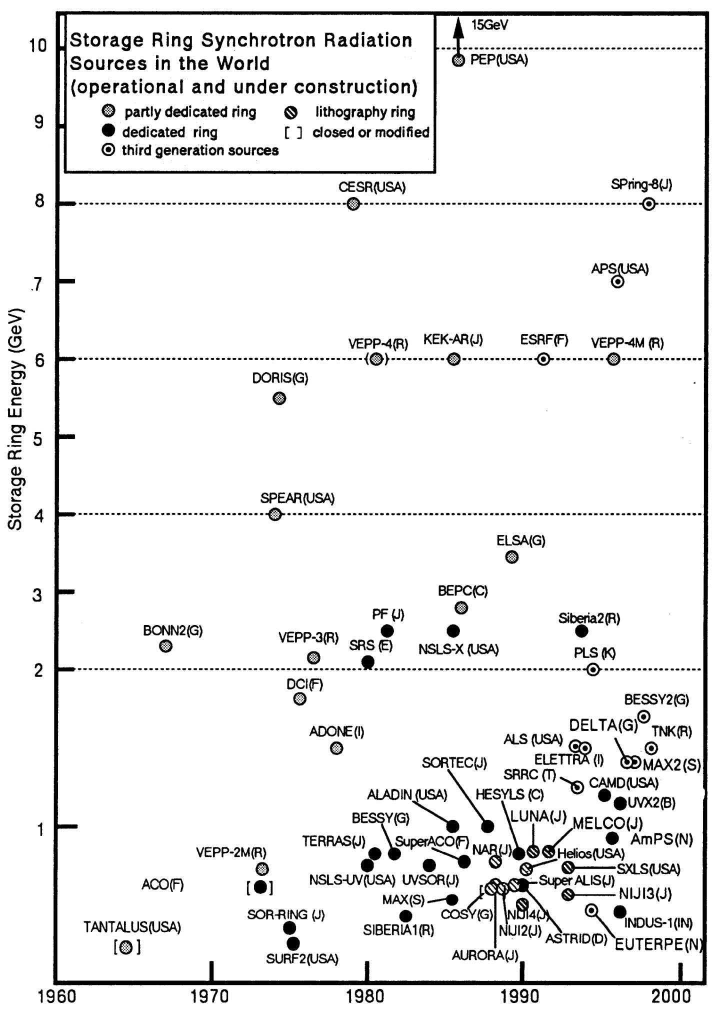
8.1.3. Insertion devices (IDs)

These are multipole magnet devices placed (inserted) in straight sections of the synchrotron or storage ring. They can be designed to enhance specific characteristics of the SR, namely

(1) to extend the spectral range to shorter wavelengths (wavelength shifter);



(*a*)



(*b*)

Fig. 8.1.2.1. (*a*) Evolution of X-ray source brilliance (photons s^{-1} $mrad^{-2}$ mm^{-2} per 0.1% $\delta\lambda/\lambda$) in the hundred years since Rontgen's discovery of X-rays in 1895. Adapted from Coppens (1992). (*b*) The evolution of storage-ring synchrotron-radiation sources over the decades, as illustrated by their increasing number and range of machine energies up to the present (Suller, 1998).

(2) to increase the available intensity (multipole wiggler);

(3) to increase the brilliance *via* interference and also yield a quasi-monochromatic beam (undulator) (Fig. 8.1.2.4*b* shows the distinctly different emission from an undulator);

(4) to provide a different polarization (*e.g.* to rotate the plane of polarization, to produce circularly polarized light *etc.*).

The classification of a periodic magnet ID as a wiggler or undulator is based on whether the angular deflection, δ, of the electron beam is small enough to allow radiation emitted from one pole to interfere directly with that from the next pole. In a wiggler, $\delta \gg \gamma^{-1}$, so the interference is negligible and the spectral emission (Fig. 8.1.2.4*a*) is very similar in shape to, but scaled up from, the universal curve (*i.e.* bending magnet spectral shape). In an undulator $\delta \leq \gamma^{-1}$ and the interference effects are highly significant (Fig. 8.1.2.4*b*). If the period of the ID is λ_u (cm), then the wavelengths λ_i (i integer) emitted are given by

$$\lambda_i = \frac{\lambda_u}{i2\gamma^2}\left(1 + \frac{K^2}{2} + \gamma^2\theta^2\right), \qquad (8.1.3.1)$$

where $K = \gamma\delta$.

The spectral width of each peak is

$$\Delta_i \simeq 1/iN, \qquad (8.1.3.2)$$

where N is the number of poles.

The angular deflection, δ, is changed by opening or closing the gap between the pole pieces. Opening the gap weakens the field and shifts the emitted lines to shorter wavelengths, but decreases the flux. Conversely, to achieve a high flux means closing the gap, and in order to avoid the fundamental emission line moving to long wavelength, the machine energy has to be high. Short-wavelength undulator emission is the province of the third-generation machines, such as the ESRF in Grenoble, France (6 GeV), the APS at Argonne National Laboratory, Chicago, USA (7 GeV), and SPring-8 at Harima Science Garden City, Japan (8 GeV). Another important consideration is to cover the entire spectral range of interest to the user *via* the tuning range of the fundamental line and harmonics. This is easier the higher the machine energy. However, important developments involving so-called narrow-gap undulators (*e.g.* from 20 mm down to $\sim$7 mm) erode the advantage of higher machine energies $\geq$6 GeV for the production of X-rays within the photon energy range of primary interest to macromolecular crystallographers, namely $\sim$30 keV down to $\sim$6 keV.

8.1.4. Beam characteristics delivered at the crystal sample

The sample acceptance, α [equation (8.1.4.1)], is a quantity to which the synchrotron machine emittance [equation (8.1.2.2)] should be matched, *i.e.*,

$$\alpha = x\eta, \qquad (8.1.4.1)$$

where x is the sample size and η the mosaic spread. For example, if $x = 0.1$ mm and $\eta = 1$ mrad (0.057°), then $\alpha = 10^{-7}$ m rad or 100 nm rad.

At the sample position, the intensity of the beam, usually focused, is a useful parameter:

$$\text{Intensity} = \text{photons per s per focal spot area.} \qquad (8.1.4.2)$$

Moreover, the horizontal and vertical convergence angles are ideally kept smaller than the mosaic spread, *e.g.* $\sim$1 mrad, so as to measure reflection intensities with optimal peak-to-background ratio.

To produce a focal spot area that is approximately the size of a typical crystal ($\sim$0.3 mm) and with a convergence angle $\sim$1 mrad

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