International Tables for Crystallography (2018). Vol. H, Section 2.5.3.2, pp. 125–128.

Considering this, Liouville's theorem given in equation (2.5.14) should be expressed as

$$S_1\alpha \leq S_2\beta. \qquad (2.5.15)$$

This states that the product of the divergence and image size can be equal to or greater than the product of the capture angle and source size. If the X-ray source is a point with zero area, the focus image from focusing optics or the cross section of a parallel beam can be any chosen size. For focusing optics, the source size must be considerably smaller than the output beam size in order to achieve a gain in flux. In this case, the flux gain is from the increased capture angle. For parallel optics, the divergence angle β is infinitely small by definition, so it is necessary to use an X-ray source as small as possible to achieve a parallel beam. Focusing optics have an advantage over parallel optics in terms of beam flux. Using an X-ray beam with a divergence much smaller than the mosaicity of the specimen crystal does not improve the resolution, but does sacrifice diffraction intensity. For many X-ray diffraction applications with polycrystalline materials, a large crossfire is acceptable as long as the diffraction peaks concerned can be resolved. The improved peak profile and counting statistics can most often compensate for the peak broadening due to large crossfire.

2.5.3.1.3. *X-ray source*

A variety of X-ray sources, from sealed X-ray tubes and rotating-anode generators to synchrotron radiation, can be used for 2D powder diffraction. The history and principles of X-ray generation can be found in many references (Klug & Alexander, 1974; Cullity, 1978). The X-ray beam intensity depends on the X-ray optics, the focal-spot brightness and the focal-spot profile. The focal-spot brightness is determined by the maximum target loading per unit area of the focal spot, also referred to as the specific loading. A microfocus sealed tube (Bloomer & Arndt, 1999; Wiesmann *et al.*, 2007), which has a very small focal spot size (10–50 µm), can deliver a brilliance as much as one to two orders of magnitude higher than a conventional fine-focus sealed tube. The tube, which is also called a 'microsource', is typically air cooled because the X-ray generator power is less than 50 W. The X-ray optics for a microsource, either a multilayer mirror or a polycapillary, are typically mounted very close to the focal spot so as to maximize the gain on the capture angle. A microsource is highly suitable for 2D-XRD because of its spot focus and high brilliance.

If the X-rays used for diffraction have a wavelength slightly shorter than the *K* absorption edge of the sample material, a significant amount of fluorescent radiation is produced, which spreads over the diffraction pattern as a high background. In a conventional diffractometer with a point detector, the fluorescent background can be mostly removed by either a receiving monochromator mounted in front of the detector or by using a point detector with sufficient energy resolution. However, it is impossible to add a monochromator in front of a 2D detector and most area detectors have insufficient energy resolution. In order to avoid intense fluorescence, the wavelength of the X-ray-tube $K\alpha$ line should either be longer than the *K* absorption edge of the sample or far away from the *K* absorption edge. For example, Cu $K\alpha$ should not be used for samples containing significant amounts of the elements iron or cobalt. Since the $K\alpha$ line of an element cannot excite fluorescence of the same element, it is safe to use an anode of the same metallic element as the sample if the X-ray tube is available, for instance Co $K\alpha$ for Co samples. In general, intense fluorescence is produced when the atomic number of the anode material is 2, 3, or 4 larger than that of an element in the sample. When the sample contains Co, Fe or Mn (or Ni or Cu), the use of Cu $K\alpha$ radiation should be avoided; similarly, one should avoid using Co $K\alpha$ radiation if the sample contains Mn, Cr or V, and avoid using Cr $K\alpha$ radiation if the sample contains Ti, Sc or Ca. The effect is reduced when the atomic-number difference increases.

2.5.3.1.4. *X-ray optics*

The function of the X-ray optics is to condition the primary X-ray beam into the required wavelength, beam focus size, beam profile and divergence. Since the secondary beam path in a 2D-XRD system is an open space, almost all X-ray optics components are on the primary side. The X-ray optics components commonly used for 2D-XRD systems include a β-filter, a crystal monochromator, a pinhole collimator, cross-coupled multilayer mirrors, a Montel mirror, a polycapillary and a monocapillary. Detailed descriptions of these optic devices can be found in Chapter 2.1. In principle, the cross-sectional shape of the X-ray beam used in a 2D diffraction system should be small and round. In data-analysis algorithms, the beam size is typically considered to be a point. In practice, the beam cross section can be either round, square or another shape with a limited size. Such an X-ray beam is typically collimated or conditioned by the X-ray optics in two perpendicular directions, so that the X-ray optics used for the point beam are often called 'two-dimensional X-ray optics'.

A pinhole collimator is normally used to control the beam size and divergence in addition to other optic devices. The choice of beam size is often a trade-off between intensity and the ability to illuminate small regions or resolve closely spaced sample features. Smaller beam sizes, such as 50 µm and 100 µm, are preferred for microdiffraction and large beam sizes, such as 0.5 mm or 1 mm, are typically used for quantitative analysis, or texture or crystallinity measurements. In the case of quantitative analysis and texture measurements, using too small a collimator can actually be a detriment, causing poor grain-sampling statistics. The smaller the collimator, the longer the data-collection time. The beam divergence is typically determined by both the collimator and the coupling optic device. Lower divergence is typically associated with a long beam path. At the same time, the beam flux is inversely proportional to the square of the distance between the source and the sample. There are two main factors determining the length of the primary beam path: the first is the required distance for collimating the beam into the required divergence, the second is the space required for the primary X-ray optics, the sample stage and the detector. On the condition that the above two factors are satisfied, the primary X-ray beam path should be kept as short as possible.

2.5.3.2. *2D detector*

Two-dimensional (2D) detectors, also referred to as area detectors, are the core of 2D-XRD. The advances in area-detector technologies have inspired applications both in X-ray imaging and X-ray diffraction. A 2D detector contains a two-dimensional array of detection elements which typically have identical shape, size and characteristics. A 2D detector can simultaneously measure both dimensions of the two-dimensional distribution of the diffracted X-rays. Therefore, a 2D detector may also be referred to as an X-ray camera or imager. There are many technologies for area detectors (Arndt, 1986; Krause & Phillips, 1992; Eatough *et al.*, 1997; Giomatartis, 1998; Westbrook, 1999; Durst *et al.*, 2002; Blanton, 2003; Khazins *et al.*, 2004). X-ray photographic plates and films were the first generation of two-dimensional X-ray detectors. Now, multiwire proportional counters (MWPCs), image plates (IPs), charge-coupled devices (CCDs) and microgap detectors are the most commonly used large area detectors. Recent developments in area detectors include X-ray pixel array detectors (PADs), silicon drift diodes (SDDs) and complementary metal-oxide semiconductor (CMOS) detectors (Ercan *et al.*, 2006; Lutz, 2006; Yagi & Inoue, 2007; He *et al.*, 2011). Each detector type has its advantages over the other types. In order to make the right choice of area detector for a 2D-XRD system and applications, it is necessary to characterize area detectors with consistent and comparable parameters. Chapter 2.1 has more comprehensive coverage on X-ray detectors, including area detectors. This section will cover the characteristics specifically relevant to area detectors.

2.5.3.2.1. *Active area and pixel size*

A 2D detector has a limited detection surface and the detection surface can be spherical, cylindrical or flat. The detection-surface shape is also determined by the detector technology. For example, a CCD detector is made from a large semiconductor wafer, so that only a flat CCD is available, while an image plate is flexible so that it is easily bent to a cylindrical shape. The area of the detection surface, also referred to as the active area, is one of the most important parameters of a 2D detector. The larger the active area of a detector, the larger the solid angle that can be covered at the same sample-to-detector distance. This is especially important when the instrumentation or sample size forbid a short sample-to-detector distance. The active area is also limited by the detector technology. For instance, the active area of a CCD detector is limited by the semiconductor wafer size and fabrication facility. A large active area can be achieved by using a large demagnification optical lens or fibre-optical lens. Stacking several CCD chips side-by-side to build a so-called mosaic CCD detector is another way to achieve a large active area.

In addition to the active area, the overall weight and dimensions are also very important factors in the performance of a 2D detector. The weight of the detector has to be supported by the goniometer, so a heavy detector means high demands on the size and power of the goniometer. In a vertical configuration, a heavy detector also requires a heavy counterweight to balance the driving gear. The overall dimensions of a 2D detector include the height, width and depth. These dimensions determine the manoeuvrability of the detector within a diffractometer, especially when a diffractometer is loaded with many accessories, such as a video microscope and sample-loading mechanism. Another important parameter of a 2D detector that tends to be ignored by most users is the blank margin surrounding the active area of the detector. Fig. 2.5.10 shows the relationship between the maximum measurable 2θ angle and the detector blank margin. For high 2θ angle measurements, the detector swing angle is set so that the incident X-ray optics are set as closely as possible to the detector. The unmeasurable blank angle is the sum of the detector margin m and the dimension from the incident X-ray beam to the outer surface of the optic device h. The maximum measurable angle is given by

$$2\theta_{\max} = \pi - \frac{m+h}{D}. \qquad (2.5.16)$$

It can be seen that either reducing the detector blank margin or optics blank margin can increase the maximum measurable angle.

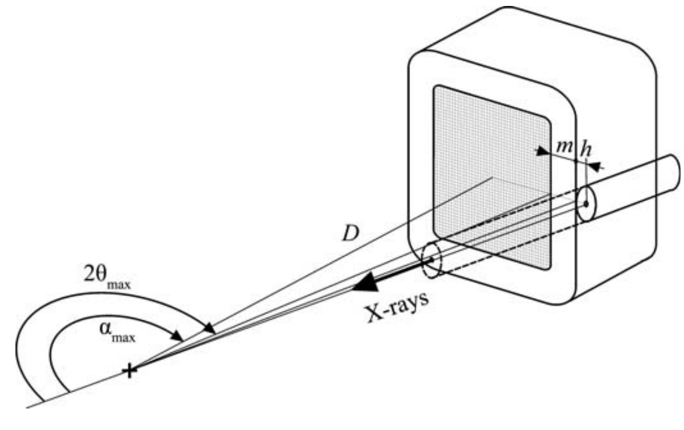

Figure 2.5.10
Detector dimensions and maximum measurable 2θ.

The solid angle covered by a pixel in a flat detector is dependent on the sample-to-detector distance and the location of the pixel in the detector. Fig. 2.5.11 illustrates the relationship between the solid angle covered by a pixel and its location in a flat area detector. The symbol S may represent a sample or a calibration source at the instrument centre. The distance between the sample S and the detector is D. The distance between any arbitrary pixel $P(x, y)$ and the detector centre pixel $P(0, 0)$ is r. The pixel size is Δx and Δy (assuming $\Delta x = \Delta y$). The distance between the sample S and the pixel is R. The angular ranges covered by this pixel are $\Delta\alpha$ and $\Delta\beta$ in the x and y directions, respectively. The solid angle covered by this pixel, $\Delta\Omega$, is then given as

$$\Delta\Omega = \Delta\alpha\Delta\beta = \frac{D}{R^3}\Delta y\Delta x = \frac{D}{R^3}\Delta A, \qquad (2.5.17)$$

where $\Delta A = \Delta x\Delta y$ is the area of the pixel and R is given by

$$R = (D^2 + x^2 + y^2)^{1/2} = (D^2 + r^2)^{1/2}. \qquad (2.5.18)$$

When a homogeneous calibration source is used, the flux to a pixel at $P(x, y)$ is given as

$$F(x, y) = \Delta\Omega B = \frac{\Delta ADB}{R^3} = \frac{\Delta ADB}{(D^2 + x^2 + y^2)^{3/2}}, \qquad (2.5.19)$$

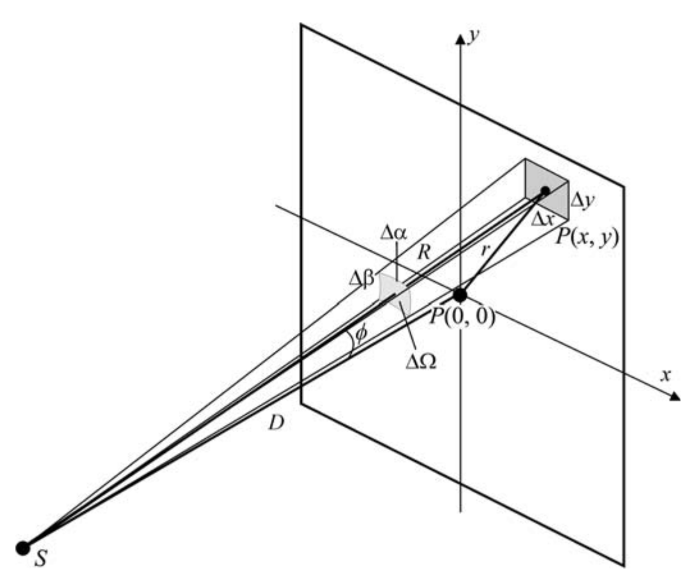

Figure 2.5.11
Solid angle covered by each pixel and its location on the detector.

where $F(x, y)$ is the flux (in photons s^{-1}) intercepted by the pixel and B is the brightness of the source (in photons s^{-1} $mrad^{-2}$) or scattering from the sample. The ratio of the flux in pixel $P(x, y)$ to that in the centre pixel $P(0, 0)$ is then given as

$$\frac{F(x, y)}{F(0, 0)} = \frac{D^3}{R^3} = \frac{D^3}{(D^2 + x^2 + y^2)^{3/2}} = \cos^3\phi, \qquad (2.5.20)$$

where ϕ is the angle between the X-rays to the pixel $P(x, y)$ and the line from S to the detector in perpendicular direction. It can be seen that the greater the sample-to-detector distance, the smaller the difference between the centre pixel and the edge pixel in terms of the flux from the homogeneous source. This is the main reason why a data frame collected at a short sample-to-detector distance has a higher contrast between the edge and centre than one collected at a long sample-to-detector distance.

2.5.3.2.2. *Spatial resolution of area detectors*

In a 2D diffraction frame, each pixel contains the X-ray intensity collected by the detector corresponding to the pixel element. The pixel size of a 2D detector can be determined by or related to the actual feature sizes of the detector structure, or artificially determined by the readout electronics or data-acquisition software. Many detector techniques allow multiple settings for variable pixel size, for instance a frame of 2048 × 2048 pixels or 512 × 512 pixels. Then the pixel size in 512 mode is 16 (4 × 4) times that of a pixel in 2048 mode. The pixel size of a 2D detector determines the space between two adjacent pixels and also the minimum angular steps in the diffraction data, therefore the pixel size is also referred to as pixel resolution.

The pixel size does not necessarily represent the true spatial resolution or the angular resolution of the detector. The resolving power of a 2D detector is also limited by its point-spread function (PSF) (Bourgeois *et al.*, 1994). The PSF is the two-dimensional response of a 2D detector to a parallel point beam smaller than one pixel. When the sharp parallel point beam strikes the detector, not only does the pixel directly hit by the beam record counts, but the surrounding pixels may also record some counts. The phenomenon is observed as if the point beam has spread over a certain region adjacent to the pixel. In other words, the PSF gives a mapping of the probability density that an X-ray photon is recorded by a pixel in the vicinity of the point where the X-ray beam hits the detector. Therefore, the PSF is also referred to as the spatial redistribution function. Fig. 2.5.12(*a*) shows the PSF produced from a parallel point beam. A plane at half the maximum intensity defines a cross-sectional region within the PSF. The FWHM can be measured at any direction crossing the centroid of the cross section. Generally, the PSF is isotropic, so the FWHMs measured in any direction should be the same.

Measuring the PSF directly by using a small parallel point beam is difficult because the small PSF spot covers a few pixels and it is hard to establish the distribution profile. Instead, the line-spread function (LSF) can be measured with a sharp line beam from a narrow slit (Ponchut, 2006). Fig. 2.5.12(*b*) is the intensity profile of the image from a sharp line beam. The LSF can be obtained by integrating the image from the line beam along the direction of the line. The FWHM of the integrated profile can be used to describe the LSF. Theoretically, LSF and PSF profiles are not equivalent, but in practice they are not distinguished and may be referenced by the detector specification interchangeably. For accurate LSF measurement, the line beam is intentionally positioned with a tilt angle from the orthogonal direction of the pixel array so that the LSF can have smaller steps in the integrated profile (Fujita *et al.*, 1992).

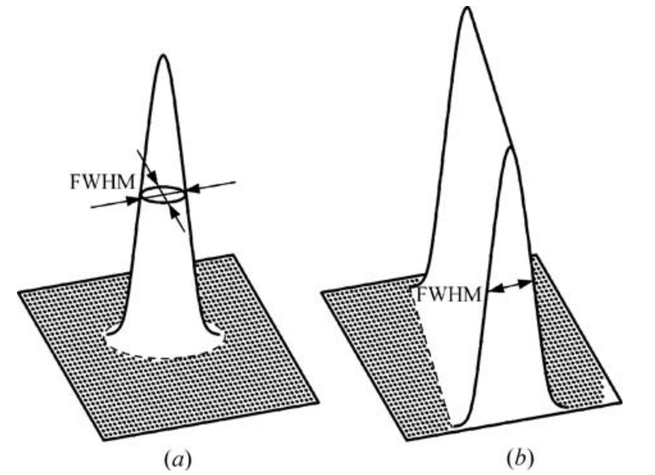

Figure 2.5.12
(*a*) Point-spread function (PSF) from a parallel point beam; (*b*) line-spread function (LSF) from a sharp line beam.

The RMS (root-mean-square) of the distribution of counts is another parameter often used to describe the PSF. The normal distribution, also called the Gaussian distribution, is the most common shape of a PSF. The RMS of a Gaussian distribution is its standard deviation, σ. Therefore, the FWHM and RMS have the following relation, assuming that the PSF has a Gaussian distribution:

$$\text{FWHM} = 2[-2\ln(1/2)]^{1/2}\text{RMS} = 2.3548 \times \text{RMS}. \qquad (2.5.21)$$

The values of the FWHM and RMS are significantly different, so it is important to be precise about which parameter is used when the value is given for a PSF.

For most area detectors, the pixel size is smaller than the FWHM of the PSF. The pixel size should be small enough that at least a 50% drop in counts from the centre of the PSF can be observed by the pixel adjacent to the centre pixel. In practice, an FWHM of 3 to 6 times the pixel size is a reasonable choice if use of a smaller pixel does not have other detrimental effects. A further reduction in pixel size does not necessarily improve the resolution. Some 2D detectors, such as pixel-array detectors, can achieve a single-pixel PSF. In this case, the spatial resolution is determined by the pixel size.

2.5.3.2.3. *Detective quantum efficiency and energy range*

The detective quantum efficiency (DQE), also referred to as the detector quantum efficiency or quantum counting efficiency, is measured by the percentage of incident photons that are converted by the detector into electrons that constitute a measurable signal. For an ideal detector, in which every X-ray photon is converted to a detectable signal without additional noise added, the DQE is 100%. The DQE of a real detector is less than 100% because not every incident X-ray photon is detected, and because there is always some detector noise. The DQE is a parameter defined as the square of the ratio of the output and input signal-to-noise ratios (SNRs) (Stanton *et al.*, 1992):

$$\text{DQE} = \left(\frac{(S/N)_{\text{out}}}{(S/N)_{\text{in}}}\right)^2. \qquad (2.5.22)$$

The DQE of a detector is affected by many variables, for example the X-ray photon energy and the counting rate. The dependence of the DQE on the X-ray photon energy defines the energy range of a detector. The DQE drops significantly if a detector is used out of its energy range. For instance, the energy range of MWPC and microgap detectors is about 3 to 15 keV. The DQE with Cu $K\alpha$ radiation (8.06 keV) is about 80%, but drops gradually when approaching the lower or higher energy limits. The energy range of imaging plates is much wider (4–48 keV). The energy range of a CCD, depending on the phosphor, covers from 5 keV up to the hard X-ray region.

2.5.3.2.4. *Detection limit and dynamic range*

The detection limit is the lowest number of counts that can be distinguished from the absence of true counts within a specified confidence level. The detection limit is estimated from the mean of the noise, the standard deviation of the noise and some confidence factor. In order to have the incoming X-ray photons counted with a reasonable statistical certainty, the counts produced by the X-ray photons should be above the detector background-noise counts.

The dynamic range is defined as the range extending from the detection limit to the maximum count measured in the same length of counting time. The linear dynamic range is the dynamic range within which the maximum counts are collected within the specified linearity. For X-ray detectors, the dynamic range most often refers to linear dynamic range, since only a diffraction pattern collected within the linear dynamic range can be correctly interpreted and analysed. When the detection limit in count rate approaches the noise rate at extended counting time, the dynamic range can be approximated by the ratio of the maximum count rate to the noise rate.

Dynamic range is very often confused with the maximum count rate, but must be distinguished. With a low noise rate, a detector can achieve a dynamic range much higher than its count rate. For example, if a detector has a maximum linear count rate of 10^5 s^{-1} with a noise rate of 10^{-3} s^{-1}, the dynamic range can approach 10^8 for an extended measurement time. The dynamic range for a 2D detector has the same definition as for a point detector, except that with a 2D detector the whole dynamic range extending from the detection limit to the maximum count can be observed from different pixels simultaneously. In order to record the entire two-dimensional diffraction pattern, it is necessary for the dynamic range of the detector to be at least the dynamic range of the diffraction pattern, which is typically in the range 10^2 to 10^6 for most applications. If the range of reflection intensities exceeds the dynamic range of the detector, then the detector will either saturate or have low-intensity patterns truncated. Therefore, it is desirable that the detector has as large a dynamic range as possible.

2.5.3.2.5. *Types of 2D detectors*

2D detectors can be classified into two broad categories: photon-counting detectors and integrating detectors (Lewis, 1994). Photon-counting area detectors can detect a single X-ray photon entering the active area. In a photon-counting detector, each X-ray photon is absorbed and converted to an electrical pulse. The number of pulses counted per unit time is proportional to the incident X-ray flux. Photon-counting detectors typically have high counting efficiency, approaching 100% at low count rate. The most commonly used photon-counting 2D detectors include MWPCs, Si-pixel arrays and microgap detectors. Integrating area detectors, also referred to as analogue X-ray imagers, record the X-ray intensity by measuring the analogue electrical signals converted from the incoming X-ray flux. The signal size of each pixel is proportional to the fluence of incident X-rays. The most commonly used integrating 2D detectors include image plates (IPs) and charge-coupled devices (CCDs).

The selection of an appropriate 2D detector depends on the X-ray diffraction application, the sample condition and the X-ray beam intensity. In addition to geometry features, such as the active area and pixel format, the most important performance characteristics of a detector are its sensitivity, dynamic range, spatial resolution and background noise. The detector type, either photon-counting or integrating, also leads to important differences in performance. Photon-counting 2D detectors typically have high counting efficiency at low count rate, while integrating 2D detectors are not so efficient at low count rate because of the relatively high noise background. An MWPC has a high DQE of about 0.8 when exposed to incoming local fluence from single photons up to about 10^3 photons s^{-1} mm^{-2}. The diffracted X-ray intensities from a polycrystalline or powder sample with a typical laboratory X-ray source fall into this fluence range. This is especially true with microdiffraction, where high sensitivity and low noise are crucial to reveal the weak diffraction pattern. Owing to the counting losses at a high count rate, the DQE of an MWPC decreases with increasing count rate and quickly saturates above 10^3 photons s^{-1} mm^{-2}. Therefore, an MWPC is not suitable for collecting strong diffraction patterns or for use with high intensity sources, such as synchrotron X-ray sources. An IP can be used in a large fluence range from 10 photons s^{-1} mm^{-2} and up with a DQE of 0.2 or lower. An IP is suitable for strong diffraction from single crystals with high-intensity X-ray sources, such as a rotating-anode generator or synchrotron X-ray source. With weak diffraction signals, the image plate cannot resolve the diffraction data near the noise floor. A CCD detector can also be used over a large X-ray fluence range from 10 photons s^{-1} mm^{-2} to very high fluence with a much higher DQE of 0.7 or higher. It is suitable for collecting diffraction of medium to strong intensity from single-crystal or polycrystalline samples. Owing to the relatively high sensitivity and high local count rate, CCDs can be used in systems with either sealed-tube X-ray sources, rotating-anode generators or synchrotron X-ray sources. With a low DQE at low fluence and the presence of dark-current noise, a CCD is not a good choice for applications with weak diffraction signals. A microgap detector has the best combination of high DQE, low noise and high count rate. It has a DQE of about 0.8 at an X-ray fluence from single photons up to about 10^5 photons s^{-1} mm^2. It is suitable for microdiffraction when high sensitivity and low noise are crucial to reveal weak diffraction patterns. It can also handle high X-ray fluence from strong diffraction patterns or be used with high-intensity sources, such as rotating-anode generators or synchrotron X-ray sources.

2.5.3.3. *Data corrections and integration*

2D diffraction patterns contain abundant information. In order to interpret and analyse 2D patterns accurately it is necessary to apply some data-treatment processes (Sulyanov *et al.*, 1994; Scheidegger *et al.*, 2000; Cervellino *et al.*, 2006; Boesecke, 2007; Rowe, 2009). Most data-treatment processes can be categorized as having one of the following four purposes: to eliminate or reduce errors caused by detector defects; to remove undesirable effects of instrument and sample geometry; to transfer a 2D frame into a format such that the data can be presented or further analysed by conventional means and software; and cosmetic treatment, such as smoothing a frame for reports and publications.

references