Support Document for the development of a Laue Suite Project aimed to develop reusable Fortran modules and user-friendly software.

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(This is a working document in which we write the basic equations and algorithms needed for the project)

(Some pictures and formulae – sections 2, 3 and 4 – presented in the document have been taken from the manual of D10 by Garry McIntyre, that are also a simplified summary of the first part of the W.R. Busing and H.A. Levy article at *Acta Cryst* (1967) **22**, 457. We follow the same conventions at those adopted in the manual of D10)

1: Simulation of single crystal data collection with 2D detectors

The purpose of the project is to write a general program (based on CrysFML) in which a visualisation of the diffraction pattern is obtained dynamically by manually orienting a crystal (using a 4C orienting device or another kind of cradle) through dials controlling the values of the orienting angles.
The single crystal diffraction pattern can be taken with monochromatic or white radiation. The crystal structure is considered to be known.

Steps

- Define completely the geometry: use the instrument structure of CrysFML.
- Define the limiting wavelengths as well as the incident spectrum in order to calculate properly the intensities
- Read the crystal structure and calculate structure factors and indices of all reflections contained in the volume of a sphere of radius equal to $2/\lambda_{min}$.
- Read the position of the detector: (γ_D, ν_D) of the flat detector centre, or γ_D of the highest angle for a banana detector. It is supposed that the dimensions of the detector surfaces are known. In the case of a banana one needs to know the angular span of the horizontally curved detector and the detector height. From this information one has to deduce the (γ_{min}, γ_{max}) and (ν_{min}, ν_{max})
- Given an orientation matrix of the crystal (or orienting manually the crystal through three angles (ϕ_x, ϕ_y, ϕ_z) fixing the crystal in the goniometric device: three dials can be created for this purpose. The value of the angles determines the orientation matrix. It is supposed that the initial position of the crystal has its Cartesian frame coincident with the fixed laboratory frame) and particular values of the orienting angles (ω,χ,ϕ) calculate the $\mathbf{z}_4$ vectors of all possible accessible reflections (d* up to $2/\lambda_{min}$)

$$\mathbf{z}_4 = (1/\lambda) \begin{bmatrix} \sin\gamma\cos\nu \\ \cos\gamma\cos\nu - 1 \\ \sin\nu \end{bmatrix} = \mathbf{\Omega}\cdot\mathbf{X}\cdot\mathbf{\Phi}\cdot\mathbf{z}_1$$

- Given the position of the 2D detector calculate all s_{1L} vectors (unitary vectors along the diffracted beams)

$$\mathbf{s}_1 = \lambda \mathbf{z}_4 + \mathbf{s}_0$$

- Determine the interception of the diffracted beam with the surface of the 2D detector calculating the coordinates and the intensity
- Make a picture of the detecting surface with all the visible spots

2: Diffractometer angles

The conventions of Busing and Levy (1967) are followed closely, but not in every detail. Much of this section has been extracted from their paper.

The instrument arrangement is illustrated schematically in Fig 1, which shows the instrument axis as vertical. Perpendicular to this axis, and passing through the instrument centre is the horizontal equatorial plane. The incident beam lies in this plane, and is directed at the sample, which is situated at the instrument centre. The detector rotates about the instrument axis to make an angle in the horizontal plane γ_m with the incident-beam direction. The crystal goniometer (an Eulerian cradle or a tilt goniometer) can be rotated independently about the same axis. Thus the χ axis, which lies in the equatorial plane, is positioned to make an angle ω with the incident-beam direction. The ϕ shaft is supported by the χ ring which permits the ϕ axis to be set at an angle χ from the vertical axis. The sample is assumed to be attached to the ϕ shaft so that it can be turned about this axis by an angle ϕ.
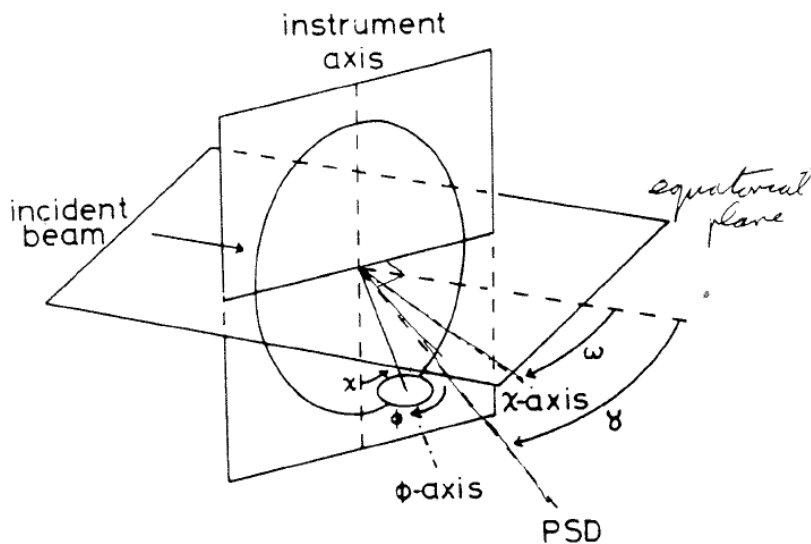

Figure 1

The diffractometer with all angles set to zero is shown schematically if Fig. 2. The senses of γ_m, ω, ϕ (all left-handed), and χ (right-handed) are defined in Fig. 1, which shows the instrument with these

angles in the first quadrant. The zero position for ϕ is chosen arbitrarily. Fig 2 also shows the laboratory system of axes x_L, y_L, z_L fixed with respect to the incident beam.
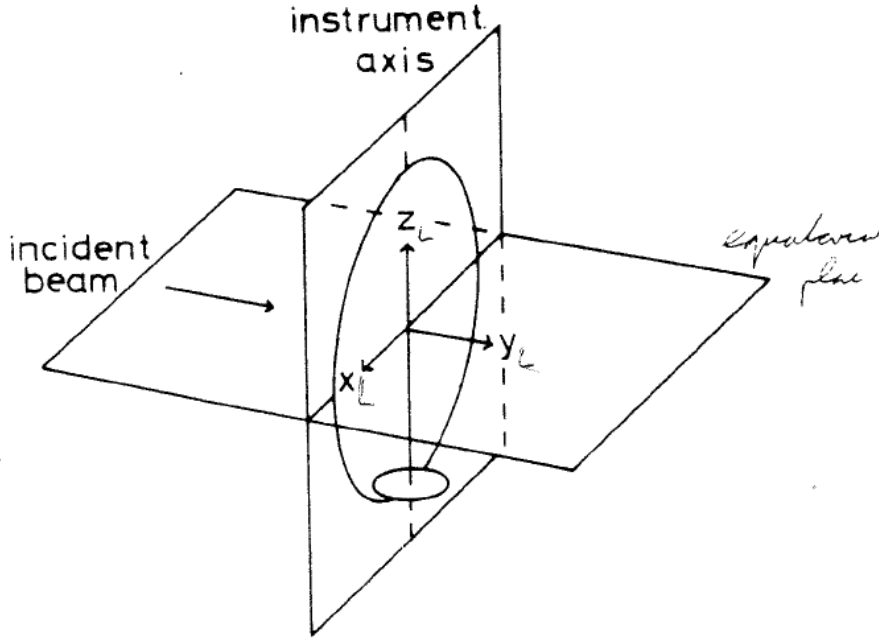

Figure 2

The angle conventions on D10 are the same as those on D9 and D19 even though the detector moves anticlockwise when viewed from above while those on D9 and D19 move anticlockwise when viewed from above. To have the same sense the laboratory axes on D10 are upside-down compared to those of D9 and D19. Thus 2θ, ω and ϕ turn clockwise for positive movements when viewed from below, and χ turns clockwise for positive movements when looking from the monochromator to the detector.

3: Instrument-coordinate transformations

Let **v** be the column vector (components v_1, v_2, v_3) describing some physical vector **v'** in terms of the reciprocal lattice vectors $\mathbf{b}_i$ (commonly denoted **a***, **b*** and **c***) so that:

$$\mathbf{v} = \sum_{i=1}^{3} v_i \mathbf{b}_i \qquad (3.1)$$

Let $\mathbf{v}_c$ be the description of such a vector in terms of the crystal Cartesian axes which are attached in some way to the reciprocal lattice. If we choose the x-axis to be parallel to $\mathbf{b}_1$, the y-axis to be in the plane of $\mathbf{b}_1$ and $\mathbf{b}_2$, and the z-axis perpendicular to that plane, then:

$$\mathbf{v}_c = \mathbf{B}\,\mathbf{v} \qquad (3.2)$$

where **B** is given by:

$$\mathbf{B} = \begin{bmatrix} b_1 & b_2 \cos \beta_3 & b_3 \cos \beta_2 \\ 0 & b_2 \sin \beta_3 & b_3 \left(\cos \beta_1 - \cos \beta_2 \cos \beta_3 \right) / \sin \beta_3 \\ 0 & 0 & b_3 \left(1 - \cos^2 \beta_1 - \cos^2 \beta_2 - \cos^2 \beta_3 + 2 \cos \beta_1 \cos \beta_2 \cos \beta_3 \right)^{1/2} / \sin \beta_3 \end{bmatrix} \qquad (3.3)$$

Here the b_i's and the β_i's are the reciprocal-lattice cell dimensions and angles respectively.

Let $\mathbf{v}_l$ be the description of a vector in terms of the laboratory system of axes, when all the instrument angles are set to zero. Let $\mathbf{U}$ be the orthogonal matrix which relates a vector expressed in the crystal Cartesian system to the description in the laboratory system when all the instrument angles are zero, so that,

$$\mathbf{v}_1 = \mathbf{U} \cdot \mathbf{v}_c \qquad (3.4)$$

$\mathbf{U}$ is called the orientation matrix since it depends on the way in which the crystal has been mounted, and also on the arc settings if a goniometer head is used.

Now suppose we rotate the crystal (and hence the vector) by the Eulerian angles ϕ, χ and ω. We wish to know the new coordinates of the vector referred to the same laboratory axes. The vector is transformed by a ϕ rotation to:

$$\mathbf{v}_2 = \mathbf{\Phi} \cdot \mathbf{v}_1 \qquad (3.5)$$

where,

$$\mathbf{\Phi} = \begin{bmatrix} \cos \phi & \sin \phi & 0 \\ -\sin \phi & \cos \phi & 0 \\ 0 & 0 & 1 \end{bmatrix}. \qquad (3.6)$$

A χ rotation will transform the vector as follows:

$$\mathbf{v}_3 = \mathbf{X} \cdot \mathbf{v}_2 \qquad (3.7)$$

where,

$$\mathbf{X} = \begin{bmatrix} \cos \chi & 0 & \sin \chi \\ 0 & 1 & 0 \\ -\sin \chi & 0 & \cos \chi \end{bmatrix}. \qquad (3.8)$$

Finally, a rotation about the ω axis will give

$$\mathbf{v}_4 = \mathbf{\Omega} \cdot \mathbf{v}_3 \qquad (3.9)$$

where

$$\mathbf{\Omega} = \begin{bmatrix} \cos\omega & \sin\omega & 0 \\ -\sin\omega & \cos\omega & 0 \\ 0 & 0 & 1 \end{bmatrix} \qquad (3.10)$$

We shall retain the use of numerical subscripts as applied here, i.e. subscript 4 for a vector in a general position oriented by ϕ, χ and ω; 3 for a vector oriented by ϕ and χ only; 2 for a vector oriented using ϕ alone; and 1 for a vector position when all angles are zero. The exception to this convention is the use of $\mathbf{s}_0$ and $\mathbf{s}_1$ to denote the incident and diffracted beams respectively.

4: The diffraction condition

If h, k and l are the Miller indices of a reflecting plane, then the corresponding column vector in the reciprocal lattice system is:

$$\mathbf{h} = \begin{bmatrix} h \\ k \\ l \end{bmatrix}. \qquad (4.1)$$
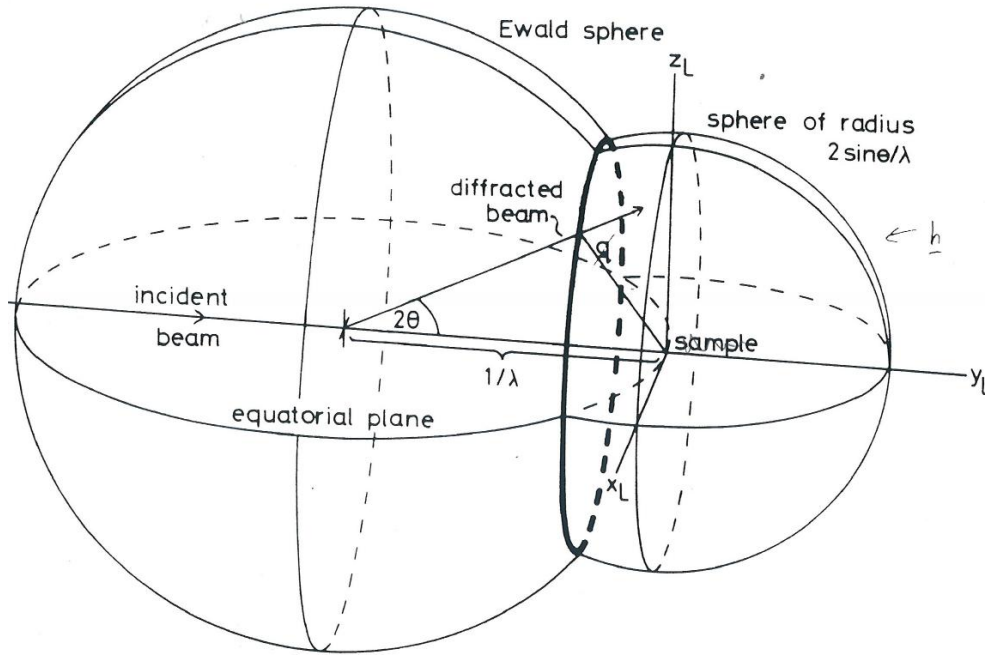

Figure 3

The length q of this vector, which is the reciprocal of interplanar spacing in Ångströms, is readily found from its components in any one of the Cartesian descriptions. For example:

$$q = \left(v_{11}^2 + v_{12}^2 + v_{13}^2 \right)^{1/2} \qquad (4.2)$$

where,

$$\mathbf{v}_1 = \mathbf{U} \cdot \mathbf{B} \cdot \mathbf{h} \qquad (4.3)$$

(We shall use the convention that v_{ij} denotes the j'th component of the vector $\mathbf{v}_i$) The Bragg equation is then,

$$\sin\theta = \lambda q / 2 \qquad (4.4)$$

In the Ewald construction of Fig. 3 the locus of the tip of the scattering vector $\mathbf{h}$ is a sphere of radius $q = 2\sin\theta/\lambda$ (λ = wavelength). The intersection of this sphere with the Ewald sphere of radius $1/\lambda$ is the bold-lined circle, called the circle of observation. Diffraction from the planes with indices hkl can occur when the tip of $\mathbf{h}$ lies at any point on this circle. Hence the instrument angles (ϕ, χ and ω) can be adjusted so that the diffracted beam occurs anywhere on the cone with semi-angle 2θ about the incident beam.

For a particular diffraction setting, $\mathbf{h}$ = OP in Fig. 4, the angular coordinates defining the diffracted beam are γ_p in the equatorial plane and ν measured vertically out of the plane, with positive ν in the direction $\mathbf{z}_L$.

The diffraction condition can be written:

$$\mathbf{h}_4 = \mathbf{S}_1 - \mathbf{S}_0 = \frac{\mathbf{S}_1 - \mathbf{S}_0}{\lambda} \qquad (4.5)$$

where $\mathbf{S}_0$ and $\mathbf{S}_1$ are vectors along the incident-beam and diffracted-beam respectively, of length $1/\lambda$ (in Fig. 4, where $\mathbf{S}_0$ = AE, and $\mathbf{S}_1$ = EP). When expanded this gives:

$$\mathbf{h}_4 = (1/\lambda) \begin{bmatrix} \sin\gamma\cos\nu \\ \cos\gamma\cos\nu - 1 \\ \sin\nu \end{bmatrix} \qquad (4.6)$$

The instrument angles which orient $\mathbf{h}$ can be solved from the basic diffractometer equation:

$$\mathbf{h}_4 = (1/\lambda) \begin{bmatrix} \sin\gamma_p\cos\nu_p \\ \cos\gamma_p\cos\nu_p - 1 \\ \sin\nu_p \end{bmatrix} = \mathbf{\Omega}\cdot\mathbf{X}\cdot\mathbf{\Phi}\cdot\mathbf{UB} \begin{bmatrix} h \\ k \\ l \end{bmatrix} \qquad (4.7)$$

The $\mathbf{U}$ and $\mathbf{B}$ matrices are usually determined together, and given as a single product $\mathbf{UB} = \mathbf{U}.\mathbf{B}$, as above.
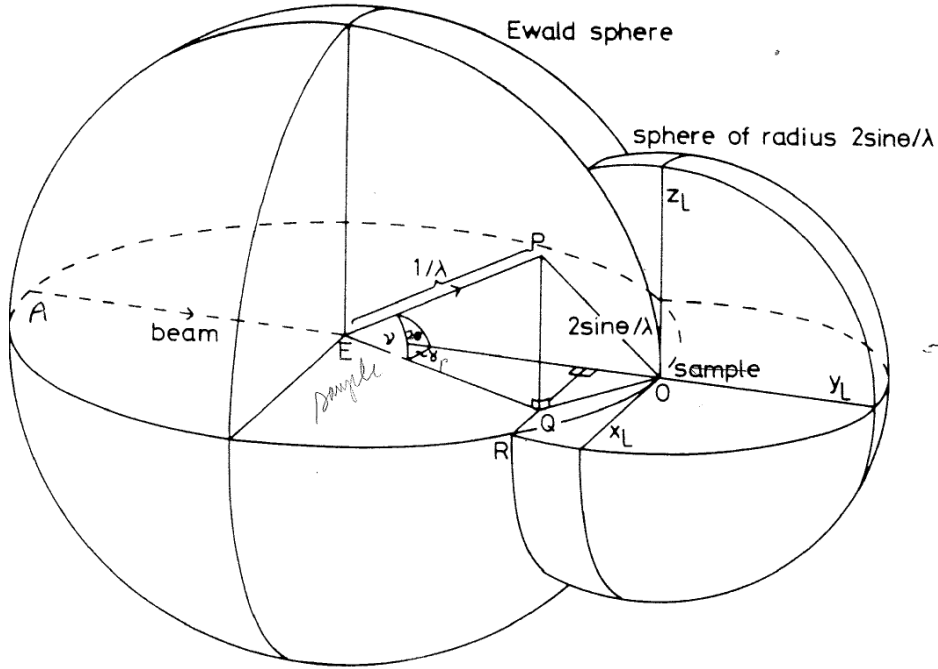
Figure 4

5: Determination of reciprocal lattice vectors referred to the Laboratory System from measured peaks for different diffractometers geometries.

If we dispose of a set of centred unknown reflections $\mathbf{h}_i$, characterized by their centres in the detector (x_i, z_i)$_D$ and the values of the angles of the orienting device we can deduce from the wavelength of the radiation and the geometry of the diffractometer a set of reciprocal vectors referred to the laboratory system $\mathbf{z}_{1i}$. These vectors are affected by errors and whatever algorithm that manipulates them in order to deduce a unit cell and an orientation matrix should take care of that.

5.1 Getting the vector z_1 from the measured peak position

Knowing the orienting angles (we suppose for the moment an Eulerian cradle) one has just to invert the following equation:

$$\mathbf{z}_4 = (1/\lambda) \begin{bmatrix} \sin\gamma\cos\nu \\ \cos\gamma\cos\nu - 1 \\ \sin\nu \end{bmatrix} = \mathbf{\Omega}\cdot\mathbf{X}\cdot\mathbf{\Phi}\cdot\mathbf{z}_1 \qquad (5.1)$$

$$\mathbf{z}_1 = \mathbf{\Phi}^{-1}\cdot\mathbf{X}^{-1}\cdot\mathbf{\Omega}^{-1}\mathbf{z}_4 = \mathbf{\Phi}^T\cdot\mathbf{X}^T\cdot\mathbf{\Omega}^T\mathbf{z}_4 \qquad (5.2)$$

The angles γ and ν can be obtained from the diffraction geometry. In the case of a limited orienting device (normal beam geometry, for instance) one can consider, for instance that we have only available the angle ϕ, or the angle ω, with $\chi=0$. Let us consider two cases a flat detector positioned by two angles γ_D and ν_D and a cylindrical detector positioned by a single arm γ_D. The geometry of diffraction is represented schematically in the following figure:
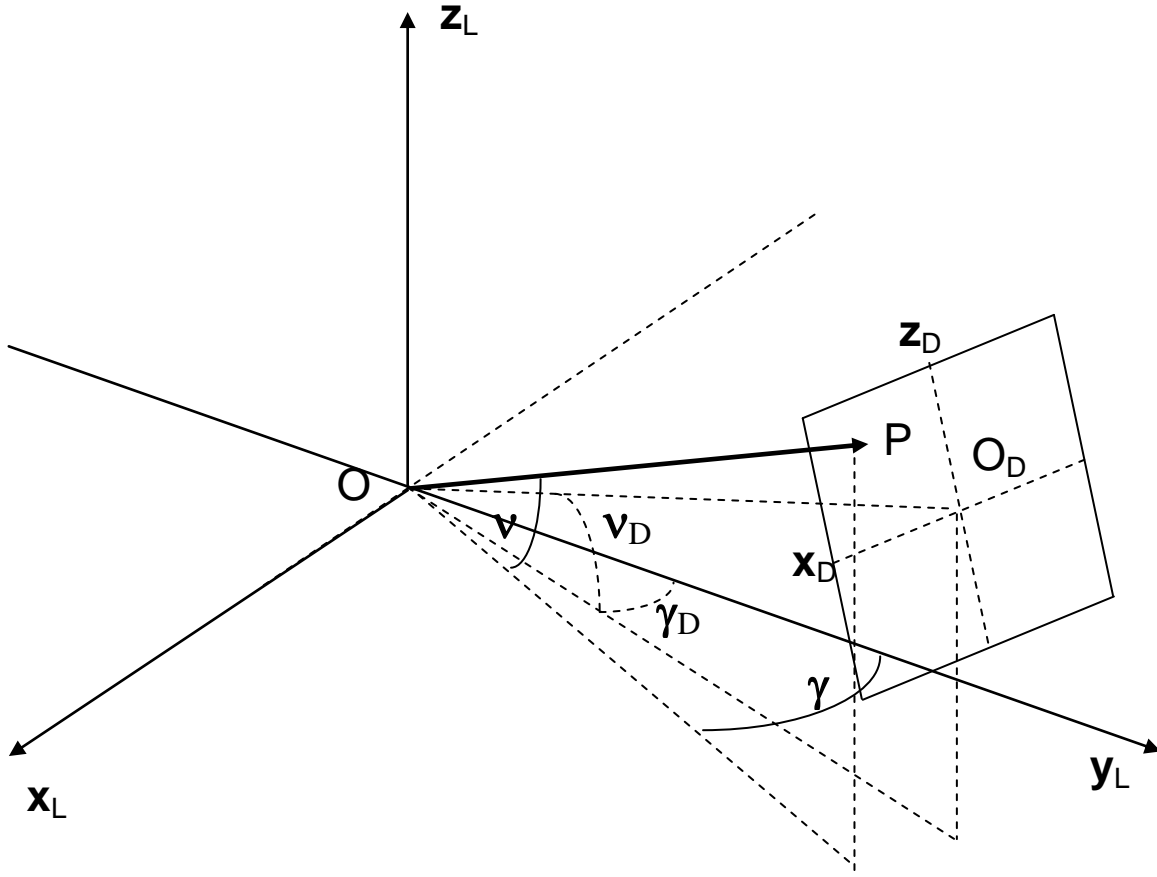
Figure 5

The vector OP, representing the diffracted beam, has as polar angles in the L-system (γ, v) These angles should be deduced from the coordinates $(x_P, z_P)_D$ and the angles (γ_D, v_D), knowing the distance $d = OO_D$ between the sample and the centre of the detector. The impact point P when the detector is at zero angles has coordinates in the laboratory system $OP_0 = (x_P, d, z_P)_L$. This point is moved to the diffraction position by applying two rotations to the vector OP_0. The first rotation is of angle v_D around $\mathbf{x}_L$ (positive: anticlockwise when seen from the positive $\mathbf{x}_L$ axis) and the second rotation is of angle γ_D around the $\mathbf{z}_L$ axis (here positive is clockwise when seen from the positive $\mathbf{z}_L$ axis), the corresponding matrices are:

$$\mathbf{N}_D = \begin{bmatrix} 1 & 0 & 0 \\ 0 & \cos v_D & -\sin v_D \\ 0 & \sin v_D & \cos v_D \end{bmatrix}; \mathbf{\Gamma}_D = \begin{bmatrix} \cos \gamma_D & \sin \gamma_D & 0 \\ -\sin \gamma_D & \cos \gamma_D & 0 \\ 0 & 0 & 1 \end{bmatrix} \tag{5.3}$$

The final components of OP in the L-system are given by:

$$\overrightarrow{OP} = \mathbf{\Gamma}_D \mathbf{N}_D \overrightarrow{OP}_0 = \begin{bmatrix} \cos \gamma_D & \sin \gamma_D & 0 \\ -\sin \gamma_D & \cos \gamma_D & 0 \\ 0 & 0 & 1 \end{bmatrix} \begin{bmatrix} 1 & 0 & 0 \\ 0 & \cos v_D & -\sin v_D \\ 0 & \sin v_D & \cos v_D \end{bmatrix} \begin{pmatrix} x_D \\ d \\ z_D \end{pmatrix} \tag{5.4}$$

The unitary vector along the diffracted beam in the laboratory system is:

$$\mathbf{s}_{1L} = (\cos v \sin \gamma, \cos v \cos \gamma, \sin v) = \frac{\overrightarrow{OP}}{OP} \tag{5.5}$$

The corresponding reflection vector in the laboratory system is:

$$\mathbf{z}_4 = \frac{\mathbf{s}_{1L} - \mathbf{s}_{0L}}{\lambda} = \frac{1}{\lambda}(\cos v \sin \gamma, \cos v \cos \gamma - 1, \sin v) \tag{5.6}$$

The modulus of the OP vector is the modulus of the vector $\mathbf{v} = (x_D, d, z_D)$, so if we define the unitary vector:

$$\mathbf{u}_0 = \frac{\mathbf{v}}{\sqrt{x_D^2 + d^2 + z_D^2}} \tag{5.7}$$

The diffraction vector $\mathbf{z}_1$ can be obtained with the matrix product:

$$\mathbf{z}_1 = \frac{1}{\lambda}\mathbf{\Phi}^T \cdot \mathbf{X}^T \cdot \mathbf{\Omega}^T \left(\mathbf{\Gamma}_D \cdot \mathbf{N}_D \mathbf{u}_0 - \mathbf{s}_{0L}\right) \tag{5.8}$$
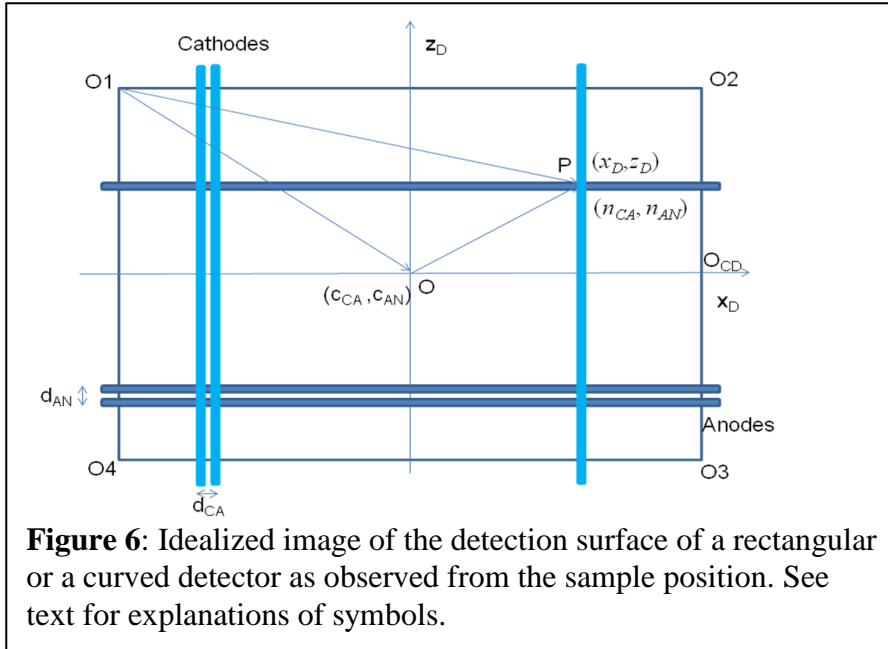
So, knowing the setting angles (ω, χ, ϕ) and the coordinates of the diffracted beam on the detector and the characteristics of the detector (distance to the sample, dimensions, curvature, etc) one can calculate the vectors $\mathbf{z}_1$ (reciprocal lattice vectors of the crystal referred to the laboratory system when all setting angles are set to zero).

If we are interested in shifts in reciprocal space and how they translate to detector space when the detector and the orienting device are fixed, the equation (5.8) simplifies as follows:

$$\delta \mathbf{z}_1 = \frac{1}{\lambda}\mathbf{\Phi}^T \cdot \mathbf{X}^T \cdot \mathbf{\Omega}^T \cdot \mathbf{\Gamma}_D \cdot \mathbf{N}_D \delta \mathbf{u}_0 \qquad \text{(Stationary detector and device)}$$

So, in terms of reciprocal space shift vectors, we obtain:

$$\delta \mathbf{u}_0 = \lambda \mathbf{N}_D^T \cdot \mathbf{\Gamma}_D^T \cdot \mathbf{\Omega} \cdot \mathbf{X} \cdot \mathbf{\Phi} \cdot \mathbf{UB} \, \delta \mathbf{h} \qquad \text{(Stationary detector and device)}$$

Figure 6: Idealized image of the detection surface of a rectangular or a curved detector as observed from the sample position. See text for explanations of symbols.

In case of a small motion of detector and orienting device, one has to calculate the differences between two expressions derived from equation (5.8). The most usual case is a coupled omega and gamma scans, so:

$$\mathbf{z}_1 - \mathbf{z}_1^a = \frac{1}{\lambda} \mathbf{\Phi}^T \cdot \mathbf{X}^T \cdot (\mathbf{\Omega}^T \mathbf{\Gamma}_D \cdot \mathbf{N}_D \mathbf{u}_0 - \mathbf{\Omega}^T \mathbf{s}_{0L} - \mathbf{\Omega}^{aT} \mathbf{\Gamma}_D^a \cdot \mathbf{N}_D \mathbf{u}_0^a - \mathbf{\Omega}^{aT} \mathbf{s}_{0L})$$

Let us make explicit some of the calculations related to different geometries and orienting devices.

5.2 Flat detector and four circles (F2D-4C)

This is the case discussed up to now. In practice the detector has its own calibration device and internal reference frame. If we consider a rectangular detector with detecting units along $\mathbf{x}_D$ that we call "cathodes", the detecting units along $\mathbf{z}_D$ are called "anodes" and we can think on that, without loss of generality, as a series of vertical (cathodes) and horizontal (anodes) wires that are numbered in some way. The coordinates of an impact point P can be provided in pixel coordinates (real numbers that become integers when the position coincide exactly with the crossing of two wires) or in "mm". For calculations we have to use length units. Passing from pixel (n_{CA}, n_{AN}) units to length units (x_D, z_D) can be performed if the starting numbering origin (O1, O2, O3, O4 in the figure), the separation between cathodes and anodes (d_{CA} and d_{AN}) and the centre of the detector (c_{CA}, c_{AN}) are known.

Suppose that we use the origin O1 (internal system) as starting point for numbering the cathodes and anodes (starting from the index 0). The x-axes coincide in direction but the z-axis of the internal system is opposite to the z-axis of the detector system.

Pixel coordinates in the internal system of the centre of detector system (c_{CA}, c_{AN})
Pixel coordinates in the internal system of the impact point P (n_{CA}, n_{AN})
Length coordinates in the internal system of the centre of detector system (X_0, Z_0)
Length coordinates in the internal system of the impact point P (X_{CA}, X_{AN})

$$\begin{aligned}(X_0, Z_0) \quad &= (d_{CA} \times c_{CA}, d_{AN} \times c_{AN}) \\ (X_{CA}, Z_{AN}) &= (d_{CA} \times n_{CA}, d_{AN} \times n_{AN}) \end{aligned} \qquad (5.9)$$

The transformation of coordinates between the internal system and the detector system is given by the following relations for the origin at O1:

$$\begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \begin{pmatrix} x_D \\ z_D \end{pmatrix} + \begin{pmatrix} X_0 \\ Z_0 \end{pmatrix} = \begin{pmatrix} X_{CA} \\ Z_{AN} \end{pmatrix}$$

$$x_D = X_{CA} - X_0; \qquad z_D = -Z_{AN} + Z_0 \qquad (5.10)$$

$$x_D = d_{CA}(n_{CA} - c_{CA}); \qquad z_D = d_{AN}(-n_{AN} + c_{AN})$$

For the case of O2 we have: $x_D = d_{CA}(-n_{CA} + c_{CA}); \qquad z_D = d_{AN}(-n_{AN} + c_{AN})$

For the case of O3 we have: $x_D = d_{CA}(-n_{CA} + c_{CA}); \qquad z_D = d_{AN}(n_{AN} - c_{AN}) \qquad (5.11)$

For the case of O4 we have: $x_D = d_{CA}(n_{CA} - c_{CA}); \qquad z_D = d_{AN}(n_{AN} - c_{AN})$

Once we have the coordinates of a spot in the detector system (x_D, z_D) and the setting angles of the detector (γ_D, ν_D) we can develop the above matrix equations and we obtain

$$\mathbf{z}_1 = \frac{1}{\lambda}\boldsymbol{\Phi}^T \cdot \mathbf{X}^T \cdot \boldsymbol{\Omega}^T \left(\boldsymbol{\Gamma}_D \cdot \mathbf{N}_D \mathbf{u}_0 - \mathbf{s}_{0L}\right) = \frac{1}{\lambda}\boldsymbol{\Phi}^T \cdot \mathbf{X}^T \cdot \boldsymbol{\Omega}^T \left(\mathbf{s}_L - \mathbf{s}_{0L}\right) \tag{5.12}$$

$$\mathbf{z}_1 = \frac{1}{\lambda}\begin{pmatrix} \cos\phi & -\sin\phi & 0 \\ \sin\phi & \cos\phi & 0 \\ 0 & 0 & 1 \end{pmatrix}\cdot\begin{pmatrix} \cos\chi & 0 & -\sin\chi \\ 0 & 1 & 0 \\ \sin\chi & 0 & \cos\chi \end{pmatrix}\cdot\begin{pmatrix} \cos\omega & -\sin\omega & 0 \\ \sin\omega & \cos\omega & 0 \\ 0 & 0 & 1 \end{pmatrix}\left(\boldsymbol{\Gamma}_D \cdot \mathbf{N}_D \mathbf{u}_0 - \mathbf{s}_{0L}\right) \tag{5.13}$$

$$\mathbf{z}_1 = \frac{1}{\lambda}\begin{pmatrix} \cos\omega\cos\chi\cos\phi - \sin\omega\sin\phi & -\sin\omega\cos\chi\cos\phi - \cos\omega\sin\phi & -\sin\chi\cos\phi \\ \cos\omega\cos\chi\sin\phi + \sin\omega\cos\phi & -\sin\omega\cos\chi\sin\phi + \cos\omega\cos\phi & -\sin\chi\sin\phi \\ \cos\omega\sin\chi & -\sin\omega\sin\chi & \cos\chi \end{pmatrix}\begin{pmatrix} n_x \\ n_y \\ n_z \end{pmatrix} \tag{5.14}$$

$$\begin{pmatrix} n_x \\ n_y \\ n_z \end{pmatrix} = \begin{pmatrix} \cos\gamma_D & \sin\gamma_D & 0 \\ -\sin\gamma_D & \cos\gamma_D & 0 \\ 0 & 0 & 1 \end{pmatrix}\begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos v_D & -\sin v_D \\ 0 & \sin v_D & \cos v_D \end{pmatrix}\begin{pmatrix} u_{0x} \\ u_{0y} \\ u_{0z} \end{pmatrix} - \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} \tag{5.15}$$

$$n_x = u_{0x}\cos\gamma_D + \sin\gamma_D(u_{0y}\cos v_D - u_{0z}\sin v_D) = \frac{1}{\sqrt{x_D^2 + d^2 + z_D^2}}\left(x_D\cos\gamma_D + \sin\gamma_D(d\cos v_D - z_D\sin v_D)\right)$$

$$n_y = -u_{0x}\sin\gamma_D + \cos\gamma_D(u_{0y}\cos v_D - u_{0z}\sin v_D) - 1 = \frac{1}{\sqrt{x_D^2 + d^2 + z_D^2}}\left(-x_D\sin\gamma_D + \cos\gamma_D(d\cos v_D - z_D\sin v_D)\right) - 1$$

$$n_z = u_{0y}\sin v_D + u_{0z}\cos v_D = \frac{1}{\sqrt{x_D^2 + d^2 + z_D^2}}\left(d\sin v_D + z_D\cos v_D\right) \tag{5.16}$$

The values (x_D, d, z_D) as well as the setting angles $(\omega, \chi, \phi, \gamma_D, v_D)$ are supposed to be the "true" ones. In fact these "true" values are obtained from the values provided by the instrument (written with an additional o-index) minus a zero-shift. The zero-shifts are common to all measured reflections and can be determined by least-squares refinement. Some of the shifts are redundant or strongly correlated. One can consider with a very good degree of approximation that only the following parameters are affected by a zero-shift:

$$x_D = X_{CA} - X_0 - \delta x_o; \qquad z_D = -Z_{AN} + Z_0 - \delta z_o; \qquad d = d_o - \delta d_o$$
$$\omega = \omega_o - \delta\omega_o; \qquad \chi = \chi_o - \delta\chi_o \tag{5.17}$$

The origin of the angle is irrelevant and the shifts on the setting angles (γ_D, v_D) of the detector when they are small can be absorbed in $(\delta x_o, \delta z_o)$. We have not considered a tilt of the detector that is generally negligible when the size of the detector is small.

In any case a series of tilts can be considered by relating the internal reference frame to the ideal detector frame through a product of three small rotations around the $\mathbf{x}_D$, $\mathbf{y}_D$ and $\mathbf{z}_D$ axes of angles (in radians) $\delta\phi_x$, $\delta\phi_y$ and $\delta\phi_z$. The rotation matrix up to first order is:

$$\mathbf{T}(\delta\phi_x, \delta\phi_y, \delta\phi_z) = \begin{pmatrix} 1 & \delta\phi_z & 0 \\ -\delta\phi_z & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}\cdot\begin{pmatrix} 1 & 0 & \delta\phi_y \\ 0 & 1 & 0 \\ -\delta\phi_y & 0 & 1 \end{pmatrix}\begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & \delta\phi_x \\ 0 & -\delta\phi_x & 1 \end{pmatrix}$$

$$\mathbf{T}(\delta\phi_x, \delta\phi_y, \delta\phi_z) = \begin{pmatrix} 1 & \delta\phi_z & \delta\phi_y \\ -\delta\phi_z & 1 & \delta\phi_x \\ -\delta\phi_y & -\delta\phi_x & 1 \end{pmatrix} \tag{5.18}$$

The simplified equation relating the cathode-anode coordinates in the form of a 2D matrix equation can be written in the general case (once the macroscopic rotation of the two frames is done) up to first order as (case O1):
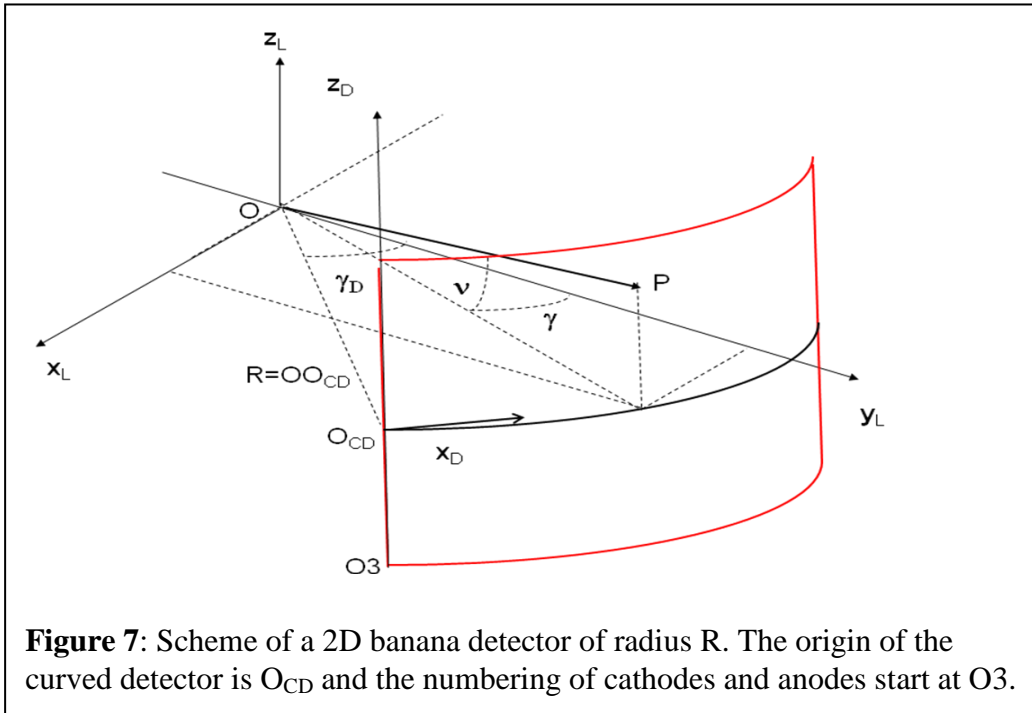
$$\begin{pmatrix} 1 & \delta\phi_z & \delta\phi_y \\ -\delta\phi_z & 1 & \delta\phi_x \\ -\delta\phi_y & -\delta\phi_x & 1 \end{pmatrix}\begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{pmatrix}\begin{pmatrix} x_D \\ y_D \\ z_D \end{pmatrix} + \begin{pmatrix} X_0 + \delta x_o \\ \delta d_o \\ Z_0 + \delta z_o \end{pmatrix} = \begin{pmatrix} X_{CA} \\ 0 \\ Z_{AN} \end{pmatrix} \qquad (5.19)$$

$$\begin{pmatrix} 1 & \delta\phi_z & \delta\phi_y \\ -\delta\phi_z & 1 & \delta\phi_x \\ -\delta\phi_y & -\delta\phi_x & -1 \end{pmatrix}\begin{pmatrix} x_D \\ y_D \\ z_D \end{pmatrix} + \begin{pmatrix} X_0 + \delta x_o \\ \delta d_o \\ Z_0 + \delta z_o \end{pmatrix} = \begin{pmatrix} X_{CA} \\ 0 \\ Z_{AN} \end{pmatrix}$$

$$\begin{pmatrix} x_D \\ y_D \\ z_D \end{pmatrix} = \begin{pmatrix} 1 & -\delta\phi_z & -\delta\phi_y \\ \delta\phi_z & 1 & -\delta\phi_x \\ \delta\phi_y & \delta\phi_x & -1 \end{pmatrix}\begin{pmatrix} X_{CA} - X_0 - \delta x_o \\ -\delta d_o \\ Z_{AN} - Z_0 - \delta z_o \end{pmatrix} = \begin{pmatrix} X_{CA} - X_0 - \delta x_o - \delta\phi_y(Z_{AN} - Z_0) \\ \delta\phi_z(X_{CA} - X_0) - \delta d_o - \delta\phi_x(Z_{AN} - Z_0) \\ \delta\phi_y(X_{CA} - X_0) - Z_{AN} + Z_0 + \delta z_o \end{pmatrix} \qquad (5.20)$$

5.3 Horizontally curved 2D-detector and four circles (C2D-4C)

The difference between this case and the previous one is that normally we are using a big banana detector so that the centre of the detector is not considered the important parameter. Looking from the sample we can see the cathodes and anodes as in the previous case for a flat detector in figure 6. The difference with the flat detector is that the distance of whatever point of the surface to the $\mathbf{z}_L$-axis is fixed and equal to the radius of the cylinder. A 3D scheme of the detector is seen in figure 7.

Figure 7: Scheme of a 2D banana detector of radius R. The origin of the curved detector is O_{CD} and the numbering of cathodes and anodes start at O3.

The z_D coordinate of a reflection in the detector is always related to the radius R and the latitude angle as:

$$\tan v = \frac{z_D}{R_D} \quad \rightarrow \quad z_D = R_D \tan v \tag{5.21}$$

On the contrary the x_D coordinate depends on the gamma arm of the banana and the origin choice. Many options can be considered. The option represented in the figure 7 provides the value of the coordinate (in a flattened picture) in mm as:

$$x_D = R_D(\gamma_D - \gamma) \tag{5.22}$$

In which the angles are expressed in radians. The relation with the internal coordinates of the detector is similar to that of a flat detector except that the separation between adjacent "cathodes" is expressed as the product of a small subtended angle by the radius of the detector: $d_{CA} = \gamma_{CA}R_D$. With the particular case of figure 7 we have:

$$x_D = X_{CA}; \qquad\qquad z_D = Z_{AN} - Z_0$$
$$x_D = d_{CA}n_{CA} = \gamma_{CA}R_D n_{CA}; \quad z_D = d_{AN}(n_{AN} - c_{AN}) \tag{5.23}$$

An important geometry relevant for Laue diffractometers is that of a complete cylindrical surface surrounding the crystal at the origin of the laboratory system with its axis coinciding with the $\mathbf{z}_L$-axis. In such a case the origin for the detector system is taken at impinging position of the transmitted beam, so that if the total surface of the detector is cut, opened and flattened, the image of the detector surface is the same as that in figure 6 in which the separation of the "cathodes" is, as before, $d_{CA} = \gamma_{CA}R_D$. The language used for referring the position of the wires is that of pixels or raster units instead of cathodes and anodes, if we replace the indices CA by H (for horizontal) and AN by V (for vertical) the expression for such a cylindrical detector, and taking the origin O1, are:

$$x_D = X_H - X_0; \qquad\qquad z_D = -Z_V + Z_0$$
$$x_D = \gamma_H R_D(n_H - c_H); \qquad z_D = d_V(-n_V + c_V) \tag{5.24}$$
$$x_D = R_D\gamma \qquad\qquad\qquad z_D = R_D \tan v$$

In the above expressions the numbers n and c are integers corresponding to horizontal and vertical pixels, the factors $\gamma_H R_D$ and d_V serve to convert to coordinates in mm.

5.4 Static flat 2D-detector in a general position and orientation with respect to the laboratory system

All the above calculations for flat detectors correspond to a flat detector that is perpendicular to the vector position going from the centre of the laboratory system to the centre of the rectangular surface. There are other cases in which the detector is rotated with respect to this position. The most general case can be considered as follows: the surface of the detector coincides with the plane $\mathbf{x}_D\mathbf{z}_D$ and the reference system of the detector (O_D, $\mathbf{x}_D$, $\mathbf{y}_D$, $\mathbf{z}_D$) coincides with the laboratory system in its initial position. The active surface is that oriented towards the incoming beam, the other side is not active. We have to take into account this fact when considering a general orientation. We consider three active rotations applied to the reference system of the detector in the following order:

1- Rotation of angle ϕ_{Dz} around the $\mathbf{z}_L$ axis: positive rotation starting from positive $\mathbf{x}_L$ going towards positive $\mathbf{y}_L$ (anti-clockwise when seen from positive $\mathbf{z}_L$ axis)
2- Rotation of angle ϕ_{Dy} around the $\mathbf{y}_L$ axis: positive rotation (anti-clockwise when seen from positive $\mathbf{y}_L$ axis)
3- Rotation of angle ϕ_{Dx} around the $\mathbf{x}_L$ axis: positive rotation (anti-clockwise when seen from positive $\mathbf{x}_L$ axis)

The total active rotation moving the detector system with respect to the laboratory system is given by the following product:

$$
\mathbf{R}_D(\phi_{Dx},\phi_{Dy},\phi_{Dz}) =
\begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos\phi_{Dx} & -\sin\phi_{Dx} \\ 0 & \sin\phi_{Dx} & \cos\phi_{Dx} \end{pmatrix}
\cdot
\begin{pmatrix} \cos\phi_{Dy} & 0 & \sin\phi_{Dy} \\ 0 & 1 & 0 \\ -\sin\phi_{Dy} & 0 & \cos\phi_{Dy} \end{pmatrix}
\cdot
\begin{pmatrix} \cos\phi_{Dz} & -\sin\phi_{Dz} & 0 \\ \sin\phi_{Dz} & \cos\phi_{Dz} & 0 \\ 0 & 0 & 1 \end{pmatrix}
\qquad (5.25)
$$

This matrix when applied to the components of a vector (in column form) in the L-system, gives the components of the transformed vector in the L-system. The columns of the matrix $\mathbf{R}_D$ represent the coordinates of the unitary vectors of the transformed reference frame (the detector frame) with respect to the L-system.

$$
\begin{pmatrix} x' \\ y' \\ z' \end{pmatrix}_L = \mathbf{R}_D \begin{pmatrix} x \\ y \\ z \end{pmatrix}_L = \begin{pmatrix} r_{11} & r_{12} & r_{13} \\ r_{21} & r_{22} & r_{23} \\ r_{31} & r_{32} & r_{33} \end{pmatrix} \begin{pmatrix} x \\ y \\ z \end{pmatrix}_L
\qquad (5.26)
$$

The transpose of this matrix, when applied to the components of a vector in the L-system provides the components of the same vector with respect to the D-system

$$
\begin{pmatrix} x \\ y \\ z \end{pmatrix}_D = \mathbf{R}_D^T \begin{pmatrix} x \\ y \\ z \end{pmatrix}_L = \begin{pmatrix} r_{11} & r_{21} & r_{31} \\ r_{12} & r_{22} & r_{32} \\ r_{13} & r_{23} & r_{33} \end{pmatrix} \begin{pmatrix} x \\ y \\ z \end{pmatrix}_L \quad \Rightarrow \quad \begin{pmatrix} x \\ y \\ z \end{pmatrix}_L = \mathbf{R}_D \begin{pmatrix} x \\ y \\ z \end{pmatrix}_D
\qquad (5.27)
$$

A vector, in the D-system, having the coordinates (x_D, y_D, z_D) has the coordinates (x_L, y_L, z_L) in the laboratory system given by the equation:

$$
\mathbf{r}_L = \mathbf{R}_D \mathbf{r}_D \Rightarrow
\begin{pmatrix} x_L \\ y_L \\ z_L \end{pmatrix} =
\begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos\phi_{Dx} & -\sin\phi_{Dx} \\ 0 & \sin\phi_{Dx} & \cos\phi_{Dx} \end{pmatrix}
\cdot
\begin{pmatrix} \cos\phi_{Dy} & 0 & \sin\phi_{Dy} \\ 0 & 1 & 0 \\ -\sin\phi_{Dy} & 0 & \cos\phi_{Dy} \end{pmatrix}
\cdot
\begin{pmatrix} \cos\phi_{Dz} & -\sin\phi_{Dz} & 0 \\ \sin\phi_{Dz} & \cos\phi_{Dz} & 0 \\ 0 & 0 & 1 \end{pmatrix}
\begin{pmatrix} x_D \\ y_D \\ z_D \end{pmatrix}
\qquad (5.28)
$$

Up to now the origin of the detector coincides with the origin of the L-system, if the origin of the detector system is translated to the position $\mathbf{r}_{OD} = (x_{OD}, y_{OD}, z_{OD})$ (coordinates in the L-system), the coordinates in the L-system of a point (x_D, y_D, z_D) in the D-system are given by:

$$
\mathbf{r}_L = \mathbf{R}_D \mathbf{r}_D + \mathbf{r}_{OD} \Rightarrow
\begin{pmatrix} x_L \\ y_L \\ z_L \end{pmatrix} =
\begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos\phi_{Dx} & -\sin\phi_{Dx} \\ 0 & \sin\phi_{Dx} & \cos\phi_{Dx} \end{pmatrix}
\cdot
\begin{pmatrix} \cos\phi_{Dy} & 0 & \sin\phi_{Dy} \\ 0 & 1 & 0 \\ -\sin\phi_{Dy} & 0 & \cos\phi_{Dy} \end{pmatrix}
\cdot
\begin{pmatrix} \cos\phi_{Dz} & -\sin\phi_{Dz} & 0 \\ \sin\phi_{Dz} & \cos\phi_{Dz} & 0 \\ 0 & 0 & 1 \end{pmatrix}
\begin{pmatrix} x_D \\ y_D \\ z_D \end{pmatrix}
+
\begin{pmatrix} x_{OD} \\ y_{OD} \\ z_{OD} \end{pmatrix}
\qquad (5.29)
$$

In some circumstances it is more convenient to use other system of rotations, for instance it may be more convenient to start with a rotation around the $\mathbf{y}_L$-axis, followed by a rotation around the $\mathbf{x}_L$-axis and finally around the $\mathbf{z}_L$-axis.

Let us consider few common special cases

a) Flat detector in forward direction, at a distance d from the sample, with the incident beam impinging the centre of the detector. In such a case $\mathbf{R}_D = \mathbf{I}$ and only the vector $\mathbf{r}_{OD} = (0, d, 0)$ is active for calculating the coordinates in the laboratory system of a spot. The point P $(x_D, 0, z_D)$ has coordinates $P_L = (x_D, d, z_D)$ in the laboratory system, so the scattering vector corresponding

to this spot is given by:

$$\mathbf{s}_L = \frac{\overrightarrow{OP}}{OP} = \frac{(x_D, d, z_D)}{\sqrt{x_D^2 + d^2 + z_D^2}} \quad \Rightarrow \quad \mathbf{z}_1 = \frac{1}{\lambda}(\mathbf{s}_L - \mathbf{s}_{0L}) = \frac{(x_D, d, z_D)}{\lambda\sqrt{x_D^2 + d^2 + z_D^2}} - (0, \frac{1}{\lambda}, 0) \quad (5.30)$$

The spherical angles (γ, ν) of the scattered beam can be obtained easily:

$$\mathbf{s}_L = (\cos\nu \sin\gamma, \cos\nu \cos\gamma, \sin\nu) = \frac{\overrightarrow{OP}}{OP} = \frac{(x_D, d, z_D)}{\sqrt{x_D^2 + d^2 + z_D^2}}$$

$$(5.31)$$

$$\nu = \arcsin\left(\frac{z_D}{\sqrt{x_D^2 + d^2 + z_D^2}}\right); \qquad \gamma = \mathrm{atan}\,2(x_D, d)$$

b) Flat detector in backward position, at a distance d from the sample, with the incident beam passing through the centre of the detector. In such a case, for detecting the spots in backward scattering, the active surface of the detector should be rotated 180° around $\mathbf{z}_L$ before translating it up to the point $\mathbf{r}_{OD} = (0, -d, 0)$

$$\mathbf{R}_D(0,0,180) = \mathbf{R}_D^T(0,0,180) = \begin{pmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \qquad (5.32)$$

The point P $(x_D, 0, z_D)$ has coordinates $P_L = (-x_D, -d, z_D)$ in the laboratory system, so the scattering vector corresponding to this spot is given by:

$$\mathbf{s}_L = \frac{\overrightarrow{OP}}{OP} = \frac{(-x_D, -d, z_D)}{\sqrt{x_D^2 + d^2 + z_D^2}} \quad \Rightarrow \quad \mathbf{z}_1 = \frac{1}{\lambda}(\mathbf{s}_L - \mathbf{s}_{0L}) = \frac{(-x_D, -d, z_D)}{\lambda\sqrt{x_D^2 + d^2 + z_D^2}} - (0, \frac{1}{\lambda}, 0) \quad (5.33)$$

The spherical angles (γ, ν) of the scattered beam can be obtained easily:

$$\mathbf{s}_L = (\cos\nu \sin\gamma, \cos\nu \cos\gamma, \sin\nu) = \frac{\overrightarrow{OP}}{OP} = \frac{(-x_D, -d, z_D)}{\sqrt{x_D^2 + d^2 + z_D^2}}$$

$$(5.34)$$

$$\nu = \arcsin\left(\frac{z_D}{\sqrt{x_D^2 + d^2 + z_D^2}}\right); \qquad \gamma = \mathrm{atan}\,2(-x_D, -d)$$

c) Flat detector with origin at position $\mathbf{r}_{OD} = (d \sin\gamma_D, d \cos\gamma_D, z_{OD})$ with $\mathbf{z}_D$ axis parallel to $\mathbf{z}_L$ and active surface perpendicular to $\mathbf{P}_D = (d \sin\gamma_D, d \cos\gamma_D, 0)$. This is the position of a detector forming part of a multidetector of a polygonal shape approximating a vertical cylindrical detector. The distance d is the distance between the two parallel axes $\mathbf{z}_D$ and $\mathbf{z}_L$. In the formalism described above this position can be reached by using the matrix:

$$\mathbf{R}_D(0,0,\phi_{Dz} = -\gamma_D) = \begin{pmatrix} \cos\gamma_D & \sin\gamma_D & 0 \\ -\sin\gamma_D & \cos\gamma_D & 0 \\ 0 & 0 & 1 \end{pmatrix} \qquad (5.35)$$

The point P $(x_D, 0, z_D)$ has coordinates in the laboratory system given by

$$\mathbf{r}_L = \mathbf{R}_D\mathbf{r}_D + \mathbf{r}_{OD} \Rightarrow \begin{pmatrix} x_L \\ y_L \\ z_L \end{pmatrix} = \begin{pmatrix} \cos\gamma_D & \sin\gamma_D & 0 \\ -\sin\gamma_D & \cos\gamma_D & 0 \\ 0 & 0 & 1 \end{pmatrix}\begin{pmatrix} x_D \\ 0 \\ z_D \end{pmatrix} + \begin{pmatrix} d\sin\gamma_D \\ d\cos\gamma_D \\ z_{OD} \end{pmatrix} = \begin{pmatrix} x_D\cos\gamma_D + d\sin\gamma_D \\ -x_D\sin\gamma_D + d\cos\gamma_D \\ z_D + z_{OD} \end{pmatrix} \qquad (5.36)$$

The spherical angles (γ, v) of the scattered beam can be easily obtained:

$$\mathbf{s}_L = (\cos v \sin \gamma, \cos v \cos \gamma, \sin v) = \frac{\mathbf{r}_L}{r_L} = \frac{(x_D \cos \gamma_D + d \sin \gamma_D, -x_D \sin \gamma_D + d \cos \gamma_D, z_D + z_{OD})}{\sqrt{(x_D \cos \gamma_D + d \sin \gamma_D)^2 + (-x_D \sin \gamma_D + d \cos \gamma_D)^2 + (z_D + z_{OD})^2}}$$

(5.37)

$$v = \arcsin\left(\frac{z_D + z_{OD}}{\sqrt{x_D^2 + d^2 + (z_D + z_{OD})^2}}\right)$$

$$\gamma = \text{atan} 2\left(x_D \cos \gamma_D + d \sin \gamma_D, \ -x_D \sin \gamma_D + d \cos \gamma_D\right)$$

d) The case shown in Figure 5 can be treated using the general formalism. The detector placed in the origin of the L-system should be rotated first an angle $\phi_{Dx} = v_D$ around $\mathbf{x}_L$-axis and then an angle $\phi_{Dz} = -\gamma_D$ around the $\mathbf{z}_L$-axis (the sense of this rotation is opposite to ϕ_{Dz}), followed to a translation to the position $\mathbf{r}_{OD} = d \ (\cos v_D \sin \gamma_D, \cos v_D \cos \gamma_D, \sin v_D)$. The point $(x_D, 0, z_D)$, with coordinates given in the detector system, has the following L-system coordinates (notice that we have emphasized the used reference frame with a subscript in the column vectors):

$$\mathbf{r}_L = \mathbf{R}_D \mathbf{r}_D + \mathbf{r}_{OD} \Rightarrow \begin{pmatrix} x_L \\ y_L \\ z_L \end{pmatrix}_L = \begin{pmatrix} \cos \gamma_D & \sin \gamma_D & 0 \\ -\sin \gamma_D & \cos \gamma_D & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos v_D & -\sin v_D \\ 0 & \sin v_D & \cos v_D \end{pmatrix} \cdot \begin{pmatrix} x_D \\ 0 \\ z_D \end{pmatrix}_D + \begin{pmatrix} x_{OD} \\ y_{OD} \\ z_{OD} \end{pmatrix}_L$$

$$\begin{pmatrix} x_L \\ y_L \\ z_L \end{pmatrix}_L = \begin{pmatrix} \cos \gamma_D & \sin \gamma_D & 0 \\ -\sin \gamma_D & \cos \gamma_D & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos v_D & -\sin v_D \\ 0 & \sin v_D & \cos v_D \end{pmatrix} \cdot \begin{pmatrix} x_D \\ 0 \\ z_D \end{pmatrix}_D + \begin{pmatrix} d \cos v_D \sin \gamma_D \\ d \cos v_D \cos \gamma_D \\ d \sin v_D \end{pmatrix}_L$$

(5.38)

$$x_L = x_D \cos \gamma_D - z_D \sin v_D \sin \gamma_D + d \cos v_D \sin \gamma_D = x_D \cos \gamma_D + \sin \gamma_D (d \cos v_D - z_D \sin v_D)$$

$$y_L = -x_D \sin \gamma_D - z_D \sin v_D \cos \gamma_D + d \cos v_D \cos \gamma_D = -x_D \sin \gamma_D + \cos \gamma_D (d \cos v_D - z_D \sin v_D)$$

$$z_L = z_D \cos \gamma_D + d \sin v_D$$

It can be easily verified that the coordinates of the point P in (5.4), obtained using another method, gives the same result:

$$\overrightarrow{OP} = \mathbf{\Gamma}_D \mathbf{N}_D \overrightarrow{OP_0} = \begin{pmatrix} \cos \gamma_D & \sin \gamma_D & 0 \\ -\sin \gamma_D & \cos \gamma_D & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos v_D & -\sin v_D \\ 0 & \sin v_D & \cos v_D \end{pmatrix} \begin{pmatrix} x_D \\ d \\ z_D \end{pmatrix}_L = \begin{pmatrix} \cos \gamma_D & \sin \gamma_D & 0 \\ -\sin \gamma_D & \cos \gamma_D & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} x_D \\ d \cos v_D - z_D \sin v_D \\ d \sin v_D + \cos v_D z_D \end{pmatrix}_L$$

$$\overrightarrow{OP} = \mathbf{r}_L = \begin{pmatrix} x_D \cos \gamma_D + \sin \gamma_D (d \cos v_D - z_D \sin v_D) \\ -x_D \sin \gamma_D + \cos \gamma_D (d \cos v_D - z_D \sin v_D) \\ d \sin v_D + z_D \cos v_D \end{pmatrix}_L$$

(5.39)

Notice that the coordinates of the vector to which the matrices are applied are given here with respect to the L-system and not with respect to the detector system.

e) Let us consider now the same case as in c) but doing first a small rotation around the $\mathbf{y}_L$-axis to the detector, The $\mathbf{R}_D$ matrix is then:

$$\mathbf{R}_D(\phi_{Dz} = -\gamma_D, \phi_{Dy} = \delta) = \begin{pmatrix} \cos \phi_{Dz} & -\sin \phi_{Dz} & 0 \\ \sin \phi_{Dz} & \cos \phi_{Dz} & 0 \\ 0 & 0 & 1 \end{pmatrix} \cdot \begin{pmatrix} \cos \phi_{Dy} & 0 & \sin \phi_{Dy} \\ 0 & 1 & 0 \\ -\sin \phi_{Dy} & 0 & \cos \phi_{Dy} \end{pmatrix} = \begin{pmatrix} \cos \gamma_D & \sin \gamma_D & 0 \\ -\sin \gamma_D & \cos \gamma_D & 0 \\ 0 & 0 & 1 \end{pmatrix} \cdot \begin{pmatrix} \cos \delta & 0 & \sin \delta \\ 0 & 1 & 0 \\ -\sin \delta & 0 & \cos \delta \end{pmatrix}$$

(5.40)

$$\mathbf{R}_D(\phi_{Dz} = -\gamma_D, \phi_{Dy} = \delta) = \begin{pmatrix} \cos \gamma_D & \sin \gamma_D & 0 \\ -\sin \gamma_D & \cos \gamma_D & 0 \\ 0 & 0 & 1 \end{pmatrix} \cdot \begin{pmatrix} 1 & 0 & \delta \\ 0 & 1 & 0 \\ -\delta & 0 & 1 \end{pmatrix} = \begin{pmatrix} \cos \gamma_D & \sin \gamma_D & \delta \cos \gamma_D \\ -\sin \gamma_D & \cos \gamma_D & -\delta \sin \gamma_D \\ -\delta & 0 & 1 \end{pmatrix}$$

We have then:

$$\mathbf{r}_L = \mathbf{R}_D \mathbf{r}_D + \mathbf{r}_{OD} \Rightarrow \begin{pmatrix} x_L \\ y_L \\ z_L \end{pmatrix} = \begin{pmatrix} \cos \gamma_D & \sin \gamma_D & \delta \cos \gamma_D \\ -\sin \gamma_D & \cos \gamma_D & -\delta \sin \gamma_D \\ -\delta & 0 & 1 \end{pmatrix} \begin{pmatrix} x_D \\ 0 \\ z_D \end{pmatrix} + \begin{pmatrix} d \sin \gamma_D \\ d \cos \gamma_D \\ z_{OD} \end{pmatrix} = \begin{pmatrix} x_D \cos \gamma_D + z_D \delta \cos \gamma_D + d \sin \gamma_D \\ -x_D \sin \gamma_D - z_D \delta \sin \gamma_D + d \cos \gamma_D \\ -x_D \delta + z_D + z_{OD} \end{pmatrix}$$ (5.41)

Let us now calculate the centre of a reflection, $\mathbf{r}_D$, in the detector reference frame. We suppose we have the following data: the rotation matrix $\mathbf{R}_D$ providing the orientation of the detector, the vector position of the centre of the detector $\mathbf{r}_{OD}$ and the polar angles (γ, v) of the diffracted beam. For calculating $\mathbf{r}_D$ we need to use the equation:

$$\mathbf{r}_D = \mathbf{R}_D^T (\mathbf{r}_L - \mathbf{r}_{OD})$$

(5.42)

We need to calculate the coordinates of the impact point in the laboratory system $\mathbf{r}_L$. For that we calculate the intersection point of the straight line with unitary vector $\mathbf{u}(\gamma, v)$ with the plane of the detector surface.

The expression of the unitary vector and the equation of the plane are:

$$\mathbf{u}(\gamma, v) = (\cos v \sin \gamma, \cos v \cos \gamma, \sin v)$$
$$(\mathbf{r}\text{-}\mathbf{r}_{OD}) \cdot \mathbf{n} = 0 \tag{5.43}$$

The normal to the plane is $\mathbf{n}=\mathbf{y}_D$ and its components with respect to the L-system constitute the second column of the matrix $\mathbf{R}_D$: $\mathbf{n}= \mathbf{R}_D(:, 2)$. The impact point $\mathbf{r}_L = p\mathbf{u}(\gamma, v)$ should satisfy the equation of the plane, this gives us the value of the distance, p, between the sample position (origin of L-system) and the impact point, we obtain:

$$(\mathbf{r}_L\text{-}\mathbf{r}_{OD}) \cdot \mathbf{n} = 0 \Rightarrow p\,\mathbf{u} \cdot \mathbf{n} = \mathbf{r}_{OD} \cdot \mathbf{n} \Rightarrow p = \frac{\mathbf{r}_{OD} \cdot \mathbf{n}}{\mathbf{u} \cdot \mathbf{n}} \Rightarrow \mathbf{r}_L = \frac{\mathbf{r}_{OD} \cdot \mathbf{n}}{\mathbf{u} \cdot \mathbf{n}}\mathbf{u} \tag{5.44}$$

The detector coordinates of the reflection spot is then given by:

$$\mathbf{r}_D = \mathbf{R}_D^T(\frac{\mathbf{r}_{OD} \cdot \mathbf{n}}{\mathbf{u} \cdot \mathbf{n}}\mathbf{u}\text{-}\mathbf{r}_{OD}) \tag{5.45}$$

For calculating the angular limits (γ, v) of the detector in an arbitrary position, we should be careful with the gamma-angles that varies in the range [-180°, 180°] with periodic boundary conditions. We have to consider separately negative gammas (γ_n) and positive gammas (γ_p). For instance, a flat detector in backscattering position have negative gamma in the range [-180°, $\gamma_n(max)$] and for positive values [$\gamma_p(min)$, 180°]. Nu-angles do not have to be considered in this way. To determine with care the angular limits, one can use a conservative approach adding and subtracting the maximum angle subtended by the vector position of the centre of the detector $\mathbf{r}_{OD} = d\,(\cos v_D \sin\gamma_D, \cos v_D \cos\gamma_D, \sin v_D)$, when the detector is perpendicular to this vector, with the vector position of one of the corner to the angles (γ_D, v_D). Once we have these angles we have to consider different options depending on the values of v_D and γ_D.

5.5 Static cylindrical 2D-detector in a general position and orientation with respect to the laboratory system

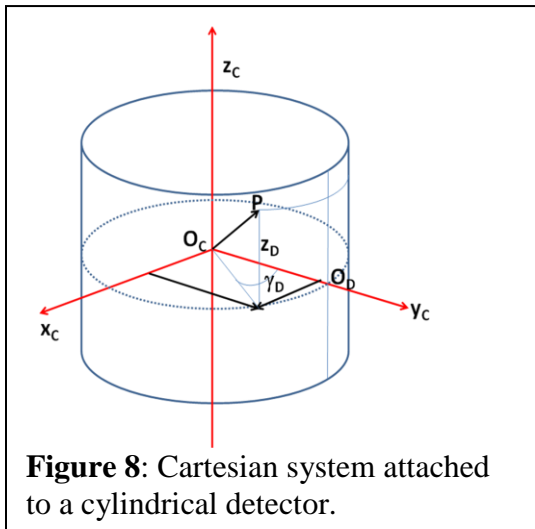**Figure 8**: Cartesian system attached to a cylindrical detector.

Let us consider a cylindrical 2D detector, o vertical dimension V(mm) and perimeter H(=2πR mm). A Cartesian coordinate system is adopted with the $\mathbf{z}_C$-axis along the cylinder axis; the origin is in the centre of the cylinder. See figure 8 for details. This Cartesian system is supposed to coincide with the L-system (this has been our implicit hypothesis in previous paragraphs referred to cylindrical detector shapes); in practice the detector may be tilted (slightly or not) and displaced with respect to the origin of the L-system. We have to find the relation between the coordinates read in the surface of the detector and the position in the L-system. The equations 5.25 to 5.29 can be used without change (except for the index C of the Cartesian system attached to the cylinder). In practice the coordinates of a spot in the detector surface (x_D, z_D),

should be transformed to the vector $O_CP=\mathbf{r}_P=(x_P, y_P, z_P)$ in order to use the equation 5.29 for obtaining the coordinates of the spot in the L-system. The coordinate z_D coincide with the coordinate z_P. The coordinate $x_D= \gamma_D R$ is the quantity provided (when transformed from pixels to mm) by the detection of peaks in the image file. The equation 5.29 with the specified notation and taking into account that we need only two tilt angles (the tilt with respect to the z-axis is immaterial) reads as:

$$\mathbf{r}_L = \mathbf{R}_D \mathbf{r}_P + \mathbf{r}_{OC} \Rightarrow \begin{pmatrix} x_L \\ y_L \\ z_L \end{pmatrix} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos\phi_{Dx} & -\sin\phi_{Dx} \\ 0 & \sin\phi_{Dx} & \cos\phi_{Dx} \end{pmatrix} \cdot \begin{pmatrix} \cos\phi_{Dy} & 0 & \sin\phi_{Dy} \\ 0 & 1 & 0 \\ -\sin\phi_{Dy} & 0 & \cos\phi_{Dy} \end{pmatrix} \begin{pmatrix} x_P \\ y_P \\ z_P \end{pmatrix} + \begin{pmatrix} x_{OC} \\ y_{OC} \\ z_{OC} \end{pmatrix} \tag{5.46}$$

The vector of a spot in the cylinder system is:

$$\mathbf{r}_P = (x_P, y_P, z_P) = (R\sin\gamma_D, R\cos\gamma_D, z_D) = (R\sin\frac{x_D}{R}, R\cos\frac{x_D}{R}, z_D) \tag{5.47}$$

The most frequent case corresponds to the inverse problem: knowing the orientation of the detector ($\mathbf{R}_D$ matrix), the vector position of the centre of the detector $\mathbf{r}_{OC}$ and the polar angles (γ, ν) of the diffracted beam, determine the spot coordinates (x_D, z_D). For calculating $\mathbf{r}_P$ we need to use the equation (5.42), which with the cylinder notation becomes:

$$\mathbf{r}_P = \mathbf{R}_D^T(\mathbf{r}_L - \mathbf{r}_{OC}) \tag{5.48}$$

The vector position of the impact point $\mathbf{r}_L = p\mathbf{u}(\gamma, \nu)$ has the unknown quantity p that has to be determined using the constraints in $\mathbf{r}_P$ (it must satisfy the equation of the cylinder in the C-system). We have the following relation that must serve to determine p: $x_P^2 + y_P^2 = R^2$. Once p determined, using the resolution of a second degree equation, the combination of equation 5.47 and 5.48 allows the determination of (x_D, z_D). Explicitly we can write the following equations:

$$\mathbf{r}_P = \mathbf{R}_D^T(\mathbf{r}_L - \mathbf{r}_{OC}) \Rightarrow (x_P, y_P, z_P) = \mathbf{R}_D^T\mathbf{r}_L - \mathbf{R}_D^T\mathbf{r}_{OC} = p\mathbf{R}_D^T\mathbf{u} - \mathbf{R}_D^T\mathbf{r}_{OC} = p\mathbf{v} - \mathbf{r}_o$$

$$(x_P, y_P, z_P) = p(v_x, v_y, v_z) - (r_{ox}, r_{oy}, r_{oz}) \Rightarrow z_P = z_D = pv_z - r_{oz}$$

$$(x_P, y_P) = p(v_x, v_y) - (r_{ox}, r_{oy}) \Rightarrow R^2 = p^2 v_{xy}^2 - 2p(v_x r_{ox} + v_y r_{oy}) + r_{oxy}^2$$

$$p^2 v_{xy}^2 - 2p(v_x r_{ox} + v_y r_{oy}) + r_{oxy}^2 - R^2 = 0 \tag{5.49}$$

From the last equation p is determined and one has to decide what of the two solutions is the adequate to the problem. This can be done looking at the signs of the (γ, ν) angles, but probably a safe way is to get the maximum value of the two solutions (e.g. $p=\max(p_1, p_2)$). With the appropriate value of p, we obtain immediately the value of the z_D coordinate (second line of eqs. 5.49). The value of x_D is obtained combining the third line of 5.49 with the equation 5.47:

$$(x_P, y_P) = (R\sin\gamma_D, R\cos\gamma_D) = (R\sin\frac{x_D}{R}, R\cos\frac{x_D}{R}) = p(v_x, v_y) - (r_{ox}, r_{oy})$$

$$\tan\frac{x_D}{R} = \frac{pv_x - r_{ox}}{pv_y - r_{oy}} \Rightarrow x_D = R\arctan\frac{pv_x - r_{ox}}{pv_y - r_{oy}} \tag{5.50}$$

6. Indexing

After an experiment for searching reflections we dispose of a set of reflections characterised by the vectors $\mathbf{z}_{1i}$ ($i = 1, 2 \ldots$ N). The method used by AJM Duisenberg (J. Appl. Cryst. 25, 92 (1992)) consists of supposing that there are various sets of lattices (due to twinning or to spurious phases) or simply several spurious reflections among the true ones. The principle of the program DIRAX is to search for one-dimensional periodicity in directions perpendicular to triplets of reflections.
(To be developed)

7. Laue patterns

For calculating a Laue pattern one can use the fact that for a fixed position of the crystal (we suppose that the orientation matrix $\mathbf{U}$ is known) there is always a set or lattice points within the two limiting Ewald spheres. Each lattice point "selects" its own wavelength between λ_{min} and λ_{max}. We dispose also of an orienting device with three angles that allows exploring all orientations of the crystal. For instance we can suppose that the crystal may be reoriented by applying three rotations like with an Eulerian cradle. The matrices corresponding to the angles can be called $\mathbf{R}(\alpha_1)$, $\mathbf{R}(\alpha_2)$, $\mathbf{R}(\alpha_3)$ and the particular expression of the matrices depends on the orienting device. The reflections that can contribute to the Laue pattern, if the orientation of the crystal can be varied at will, are those within the resolution sphere of radius $R_{res}=2/\lambda_{min}$. To obtain the Cartesian components of a reflection $\mathbf{h}$ in the laboratory frame we have:

$$\mathbf{h}_L = \mathbf{R}(\alpha_3) \cdot \mathbf{R}(\alpha_2) \cdot \mathbf{R}(\alpha_1) \cdot \mathbf{UB} \begin{bmatrix} h \\ k \\ l \end{bmatrix} = \mathbf{M} \cdot \mathbf{UB} \cdot \mathbf{h} \qquad (7.1)$$

If we consider the construction of Fig 3, the reflection $\mathbf{h}_L=(x, y, z)$ should be in the surface on one Ewald sphere corresponding to the wavelength λ. So that from the equation of the sphere in the laboratory frame we obtain:

$$x^2 + (y + \frac{1}{\lambda})^2 + z^2 = \frac{1}{\lambda^2} \quad \Rightarrow \quad x^2 + y^2 + z^2 = -\frac{2y}{\lambda}$$

$$\lambda = \frac{-2y}{x^2 + y^2 + z^2} = -2yd_{\mathbf{h}}^2 \qquad \sin\theta = -yd_{\mathbf{h}} \qquad (7.2)$$

Where $d_{\mathbf{h}}$ is the d-spacing of the reflection $\mathbf{h}$. The y coordinate of $\mathbf{h}_L$ should be negative!
If the calculated λ is outside the interval (λ_{min}, λ_{max}) the reflection will not contribute to the Laue pattern. If the reflection has the appropriate λ, the unitary vector corresponding to the diffracted beam is given by:

$$\mathbf{s}_{1L} = (\cos\nu \sin\gamma, \cos\nu \cos\gamma, \sin\nu) = \mathbf{s}_{0L} + \lambda\mathbf{h}_L = (0,1,0) + \lambda(x, y, z) \qquad (7.3)$$

The L-system coordinates of all reflections $\mathbf{h}_L=(x, y, z)$ are easily calculated from the orientation matrix and the matrices of the orienting device, the angles γ and ν are calculated from:

$$\sin\nu = \lambda z \qquad \tan\gamma = \frac{\lambda x}{1 + \lambda y} \qquad (7.4)$$

For a cylindrical detector, or radius R_D and height $2H$, surrounding the sample the γ–angle is not limited in principle (in practical cases $-\gamma_{max} < \gamma < \gamma_{max}$, with $\gamma_{max} < 180^\circ$), but the absolute value of the latitude angle v should be smaller than

$$\tan v_{max} = \frac{H}{R_D} \tag{7.5}$$

7.1 Determination of the orientation matrix using one or several Laue patterns.

In Laue diffractometers the orienting device is limited to a single angle (spindle) that corresponds to ϕ (or ω) angle (with $\chi=0$) in the Eulerian cradle, so that the general equation for a flat detector with the centre in the horizontal plane ($\mathbf{N_D=I}$):

$$\mathbf{z}_1 = \frac{1}{\lambda} \mathbf{\Phi}^T \cdot \mathbf{X}^T \cdot \mathbf{\Omega}^T \left(\mathbf{\Gamma}_D \cdot \mathbf{N}_D \mathbf{u}_0 - \mathbf{s}_{0L} \right) = \frac{1}{\lambda} \mathbf{\Phi}^T \cdot \mathbf{X}^T \cdot \mathbf{\Omega}^T \left(\mathbf{s}_L - \mathbf{s}_{0L} \right) \tag{7.6}$$

reduces to:

$$\mathbf{z}_1 = \frac{1}{\lambda} \mathbf{\Phi}^T \left(\mathbf{\Gamma}_D \mathbf{u}_0 - \mathbf{s}_{0L} \right) = \frac{1}{\lambda} \mathbf{\Phi}^T \left(\mathbf{s}_L - \mathbf{s}_{0L} \right) \tag{7.7}$$

In a Laue pattern the wavelength of the spots is not known a priori. From the coordinates of the centre of a spot we can determine only the vectors $\mathbf{z}_{Laue} = \lambda \mathbf{z}_1$, so only the directions of reciprocal vectors are directly determined. For indexing/orienting a crystal from a Laue pattern we have to know the unit cell parameters and compare the relative angles between spots and those of calculated angles of a series of generated reflections from the known cell parameters. The expression of the $\mathbf{z}_{Laue}$ vectors for a flat detector is given by:

$$\mathbf{z}_{Laue} = \lambda \mathbf{z}_1 = \begin{pmatrix} \cos\phi & -\sin\phi & 0 \\ \sin\phi & \cos\phi & 0 \\ 0 & 0 & 1 \end{pmatrix} \cdot \left(\frac{1}{\sqrt{x_D^2 + d^2 + z_D^2}} \begin{bmatrix} \cos\gamma_D & \sin\gamma_D & 0 \\ -\sin\gamma_D & \cos\gamma_D & 0 \\ 0 & 0 & 1 \end{bmatrix} \begin{pmatrix} x_D \\ d \\ z_D \end{pmatrix} - \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} \right) \tag{7.8}$$

For the case of a cylindrical detector we can apply the general equation of the direction for the scattering vector:

$$\mathbf{z}_{Laue} = \lambda \mathbf{h}_L = \mathbf{\Phi}^T \left(\mathbf{s}_L - \mathbf{s}_{0L} \right) = \begin{pmatrix} \cos\phi & -\sin\phi & 0 \\ \sin\phi & \cos\phi & 0 \\ 0 & 0 & 1 \end{pmatrix} \cdot \begin{pmatrix} \cos v \sin\gamma \\ \cos v \cos\gamma - 1 \\ \sin v \end{pmatrix} \tag{7.9}$$

remembering that

$$\gamma = \frac{x_D}{R_D} \qquad \tan v = \frac{z_D}{R_D} \tag{7.10}$$

The above equations, using raw positions of spots and the known values of spindle angle and detector, provides a list of unitary vectors that can be used to compare with calculated ones and start the indexing and consequently the determination of the orientation of the crystal.

7.2 Algorithms for indexing when the unit cell is approximately known

For obtaining the most prominent Laue spots using the experimental images one can either use a GUI for clicking in the selected spots or automatically detect the positions of spots having intensity above a prescribed threshold. For the second algorithm described below this last method is important because a

mask can be generated from the experimental image and compared with a calculated mask directly to determine the proper orientation matrix.

Indx-Algorithm 1

Step 1: Let us consider that we have a series of spots measured in a single or different Laue images. Each spot is characterized by the value of the coordinates (x, z) and spindle angle ϕ. Those with the same spindle angle belong to the same image. The user selects two or three nodal spots, put on top of the list, used for starting the procedure.

The input list is:

$$x_1, z_1, \phi_1 \; ; \; x_2, z_2, \phi_2 \; ; \; x_3, z_3, \phi_3 \; .... \; x_n, z_n, \phi_n \qquad \text{(Indx-A1-1)}$$

Step 2: From the above list, using the appropriate equations, one can calculate the Bragg angles and the vectors $\mathbf{z}_{Laue,i}$ in the laboratory system. So that:

$$\cos 2\theta_i = \cos v_i \sin \gamma_i$$
$$\mathbf{z}_{Laue,i} = \lambda \, \mathbf{UB} \cdot \mathbf{h}_i \quad (i = 1, 2, ...n) \qquad \text{(Indx-A1-2)}$$

The polar angles (γ_i, v_i) are deduced from the coordinates (x_i, z_i) and the geometry of the diffractometer. For the vectors, the known quantities are those of the left.

Step 3: Calculate the angles between the vectors pair(s): $\alpha_1 = \{\mathbf{z}_{Laue,1}, \mathbf{z}_{Laue,2}\}$, $\alpha_2 = \{\mathbf{z}_{Laue,1}, \mathbf{z}_{Laue,3}\}$, $\alpha_3 = \{\mathbf{z}_{Laue,3}, \mathbf{z}_{Laue,2}\}$

Step 4: Generate a series of nodal reflections (co-prime *hkl* indices) up to a maximum index. Calculate pair of angles that match the above ones. Calculate first two reflections that match the first angle. If solutions are found we can stop here because the orientation matrix **U** can be calculated (three free parameters) already. From the indices and the Bragg angle one can calculate the wavelength and hence the vectors $\mathbf{z}_{1,1} = \mathbf{z}_{Laue,1} / \lambda$ and $\mathbf{z}_{1,2} = \mathbf{z}_{Laue,2} / \lambda$. Now the **U** matrix can be calculated using the algorithm proposed by Busing and Levy for obtaining an orthogonal matrix.

One can continue looking for another reflection in order to be sure that the good solution is found. From the list of possible solutions (satisfying the first angular constraint) search a third reflection in the rest of generated reflections that satisfy the two additional angles. If the reflection is found we can proceed as before but with a more reliable solution.

Step 5: With the solution(s) found, calculate the indices of the rest of given reflections in the list.

Step 6: Once an initial orientation matrix is determined one can use the whole set of reflections for refining the orientation matrix, the unit cell (maintaining constant the volume) the shift, tilts and distortion parameters using least-squares.

Indx-Algorithm 2

This algorithm is based in the attribution of indices to a single noticeable nodal spot in a single image. It is always supposed that the unit cell is already known.

Step 1: The list of the most intense spots in a single image is provided. The input list is:

$$x_1, z_1; \quad x_2, z_2; \quad x_3, z_3 \ldots \ldots \quad x_n, z_n$$

Step 2: The prominent nodal spot is selected (x_p, z_p) and low indices are attributed. The unitary reciprocal vector $\mathbf{u} = \mathbf{z}_{Laue,p}/|\mathbf{z}_{Laue,p}|$ is calculated. The Bragg angle and wavelength are calculated.

Step 3: An appropriate rotation of the whole set of reflections, in the standard orientation of the crystal: the laboratory system coincides with the internal Cartesian frame of the crystal, is performed in order to put the direction of the prominent nodal spot coincident with the experimental one. The spherical angles of $\mathbf{u}_{st}$ are calculated and a rotation matrix putting $\mathbf{u}_{st}$ in coincidence with $\mathbf{u}$ is derived. We have determined here two of the three angles determining the orientation matrix $\mathbf{U}$.

Step 4: We make simulations of the Laue pattern obtained by rotating the crystal in such a way as maintaining the prominent spot fixed in the experimental position. The correct orientation is obtained when the correlation index is maximal. One can calculate this correlation index as the sum of the product of the experimental image mask (a matrix with zeros everywhere except in pixels close to the Laue spots with values equal to 1) with a calculated mask according to the calculated position of Laue spots.

Indx-Algorithm 3

This algorithm is a variant of the previous one but many images can be used simultaneously. It is always supposed that the unit cell is already known.

Step 1: As in algorithm 1

Step 2: As in algorithm 1

Step 3: From the list of vectors $\mathbf{z}_{Laue,i}$ (i=1,2 ... n), calculate the observed "star" of unitary vectors:
$$\{\mathbf{u}_o\} : \{\mathbf{u}_{oi} = \mathbf{z}_{Laue,i}/|\mathbf{z}_{Laue,i}| \quad (i = 1, 2, \ldots n)\} \qquad \text{(Indx-A3-1)}$$

Step 4: Calculate the spherical angles (θ_{oi}, ϕ_{oi}) of these vectors and construct a rectangular matrix indexed with a coarse grid on spherical angles and equal to zero everywhere except at angular positions closest to the observed peaks are, for which the value are set to 1. For fixing ideas we can consider angular steps of 1 degree, in such a case and disregarding instrumental angular limitations, the matrix dimension are (0:179, 0:359) and let us call the set of observed angles $\{\theta, \phi\}_o$ in which we have taken the near integer angle to the observed ones. The observed angular matrix is then defined as:
$$\mathbf{A}_o : \{A_{o,ij} = 0 \,|\, if \quad (i, j) \notin \{\theta, \phi\}_o; \quad A_{o,ij} = 1 \,|\, if \quad (i, j) \in \{\theta, \phi\}_o\} \qquad \text{(Indx-A3-2)}$$

Step 5: Generate a series of nodal reflection vectors (co-prime *hkl* indices) up to a given maximum index. Calculate the Cartesian components with respect to the L-system of these reflections for the standard orientation (Cartesian crystallographic system coincident with the L-system). Calculate the corresponding "star" of unitary vectors for the standard orientation:
$$\{\mathbf{u}_s\} : \{\mathbf{u}_{s,j} = \mathbf{z}_{1,j}/|\mathbf{z}_{1,j}| \quad (j = 1, 2, \ldots m)\} \qquad \text{(Indx-A3-3)}$$

and the corresponding spherical angles $\{\theta,\phi\}_s = \{\theta_{s,j},\phi_{s,j} \mid j=1,2 \ldots m\}$. Different orientations can be obtained by systematic applying a common rotation matrix to the above set of unitary vectors that can be compared with the observed set.

Step 6: Generate a rotation matrix $R_{\Theta\Phi}$ corresponding to the spherical angles $\{\Theta,\Phi\}_R$ obtained from a first rotation around the $\mathbf{y}_L$-axis of angle Θ followed by a rotation of angle Φ around the $\mathbf{z}_L$-axis. When applied to the unitary vector $\{\mathbf{u}_s\}$, the matrix $R_{\Theta\Phi}$ provides a new orientation of the crystal. This is equivalent to changing all the calculated spherical angles as follows:

$$R_{\Theta\Phi}\{\theta_{s,j},\phi_{s,j}\} = \{\theta_{s,j}+\Theta,\phi_{s,j}+\Phi\} \quad (j=1,2,\ldots m) \qquad \text{(Indx-A3-4)}$$

From this set of spherical angles we can obtain a calculated angular matrix $\mathbf{A}_c(\Theta,\Phi)$ with the same procedure, using the same dimensions, as in step **4**. The action of the rotation matrix $R_{\Theta\Phi}$ on the star $\{\mathbf{u}_s\}$ is then equivalent to generate an angular matrix $\mathbf{A}_c(\Theta,\Phi)$.

Step 7: Once the angular matrix has been constructed the correlation coefficient between the observed and calculated matrices (using a step of 1 degree) is calculated as:

$$r = \frac{\displaystyle\sum_{\theta=0,\phi=0}^{\theta=179,\phi=359} \mathbf{A}_{o,\theta\phi}\mathbf{A}_{c,\theta\phi}(\Theta,\Phi)}{\displaystyle\sum_{\theta=0,\phi=0}^{\theta=179,\phi=359} \mathbf{A}_{o,\theta\phi} \sum_{\theta=0,\phi=0}^{\theta=179,\phi=359} \mathbf{A}_{c,\theta\phi}(\Theta,\Phi)} \qquad \text{(Indx-A3-5)}$$

The obtained value is compared with a previous calculated value, r_{max}, and if it is greater the values $(\Theta,\Phi)_o$ are stored together with the new maximum r_{max}.

Step 8: The steps 6 to 7 are repeated until a pre-selected grid of (Θ,Φ) is exhausted. Or a maximisation procedure is applied. The orientation of the crystal with respect to the laboratory system is obtained at the end, so the orientation matrix corresponds to r_{max} and $(\Theta,\Phi)_o$.

$$\mathbf{U} = R_{(\Theta_o\Phi_o)} = \begin{pmatrix} \cos\Phi_o & \sin\Phi_o & 0 \\ -\sin\Phi_o & \cos\Phi_o & 0 \\ 0 & 0 & 1 \end{pmatrix}\begin{pmatrix} \cos\Theta_o & 0 & \sin\Theta_o \\ 0 & 1 & 0 \\ -\sin\Theta_o & 0 & \cos\Theta_o \end{pmatrix} \qquad \text{(Indx-A3-6)}$$

With this orientation matrix we can attribute indices to the observed reflections and multiplying by the **B**-matrix, the **UB** matrix is determined.

Appendix A

Detection of reflections in diffraction position for a monochromatic diffractometer equipped with a 2D detector

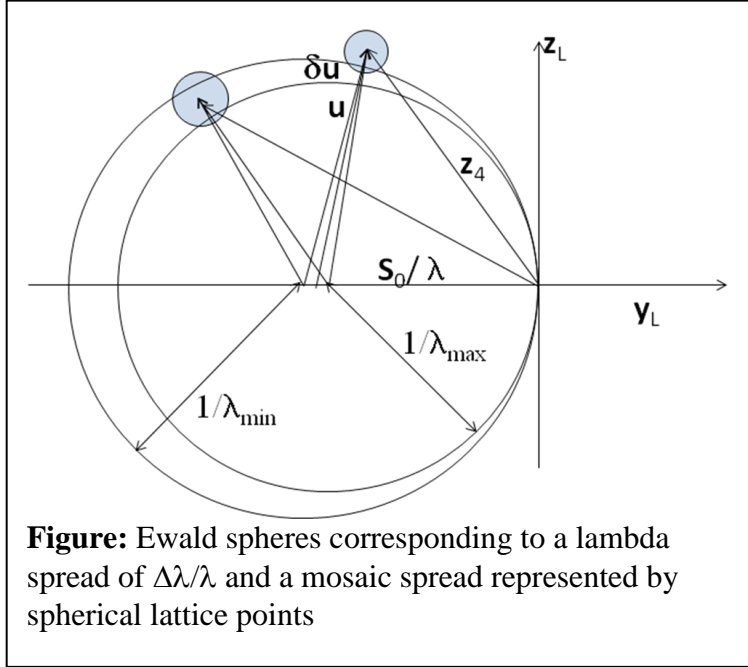We consider a simplified model of mosaicity for which each lattice point of the reciprocal space is a sphere of radius $r = 1/2\ \eta\ s$, where η is the mosaic spread expressed in radians and s is the modulus of the reciprocal vector $\mathbf{s}$. The wavelength spread is in the interval between $\lambda_{min}=\lambda-\Delta\lambda$ and $\lambda_{max}=\lambda+\Delta\lambda$. In terms of $x=\Delta\lambda/\lambda$ the interval is $[\lambda(1-x), \lambda(1+x)]$. The horizontal divergence of the incoming beam can be represented by a small rotation around the $\mathbf{z}_L$ axis of $\pm\alpha_h$ and the vertical divergence by a rotation of $\pm\alpha_v$ around the $\mathbf{x}_L$ axis. For a reciprocal lattice vector that, in Cartesian components with respect to the laboratory system, is denoted by the vector $\mathbf{z}_4=\Omega\ X\ \Phi\ \mathbf{z}_1$, a beam can be detected if the diffraction domain intersect the Ewald spheres at some point. See the above figure to see the effect of mosaic spread and wavelength dispersion. The

Figure: Ewald spheres corresponding to a lambda spread of $\Delta\lambda/\lambda$ and a mosaic spread represented by spherical lattice points

conditions for getting some intensity diffracted by the lattice point $\mathbf{z}_4$, are easily deduced from the figure:

$$\mathbf{z}_4 = \Omega X \Phi\, \mathbf{z}_1 = \Omega X \Phi\, \mathbf{UB}\,\mathbf{h}$$

$$\mathbf{v} = \frac{\mathbf{S}_0}{\lambda} + \mathbf{z}_4 = \mathbf{u} + \delta\mathbf{u}$$

$$\mathbf{u} = \frac{\mathbf{v}}{v\lambda} \Rightarrow \delta\mathbf{u} = \mathbf{v}(1 - \frac{1}{v\lambda})$$

The vector $\delta\mathbf{u}$ is along the direction of $\mathbf{v}$ joining the centre of the Ewald sphere to the centre of the lattice point. It joints a point of the surface of the Ewald sphere ($\mathbf{u}$-vector) to the centre of the lattice point. We will have diffracted intensity if the condition:

$$\text{C1:} \qquad |\delta\mathbf{u}\,| < 1/2\ \eta\ |\mathbf{z}_4|$$

is satisfied. The $\delta\mathbf{u}$ vectors can be calculated for the central wavelength, for λ_{min} and for λ_{max}. If for some of these calculations we satisfy C1 there is diffracted intensity.

We can take into account the effect of horizontal and vertical divergence by applying the condition C1 also to the eight vectors:

$R_z(\alpha_h)\mathbf{z}_4,\ R_z(-\alpha_h)\mathbf{z}_4,\ R_x(\alpha_v)\mathbf{z}_4,\ R_x(-\alpha_v)\mathbf{z}_4,\ R_x(-\alpha_v)R_z(-\alpha_h)\mathbf{z}_4,\ R_x(\alpha_v)R_z(-\alpha_h)\mathbf{z}_4$
$R_x(-\alpha_v)R_z(\alpha_h)\mathbf{z}_4,\ R_x(\alpha_v)R_z(\alpha_h)\mathbf{z}_4$

A more simple calculation can be done by considering an average divergence α that may be added quadratically to the mosaic spread. This provides an effective mosaic spread:

$$\eta_{eff} = \sqrt{\eta^2 + \alpha^2}$$

The condition C1 may be satisfied by more than a single vector, depending on the purpose of the calculations we can label the different conditions if required. For calculating the characteristics of the diffracted beam we have to use the reciprocal vector:

$$\mathbf{s} = \mathbf{u} - \frac{\mathbf{S}_0}{\lambda}$$

that can be calculated from the equations above.