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Angle calculations for a '4S+2D' six-circle diffractometer

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Abstract

Angle calculations are derived for the operation of a '4S+2D' six-circle X-ray or neutron diffractometer. The six degrees of freedom [four sample-orienting ('4S') and two detector-orienting ('2D')] of the diffractometer are used to control the scattering vector and a reference vector in the laboratory frame of reference. Several modes of operation unavailable in other types of diffractometer are presented.

1. Introduction

In recent years, several types of diffractometer have been developed for the study of single crystals with monochromatic X-rays or neutrons. One of the most popular instruments is the four-circle diffractometer, which consists of one detector circle and three sample-orienting circles. Since only three degrees of freedom are needed to position a sample crystal in an orientation satisfying a desired Bragg condition, the extra degree of freedom in the crystal orienting circles is used in various modes of diffractometer operation, typically for scattering measurements of bulk single-crystal samples. Angle calculation schemes for the four-circle diffractometer were developed by Busing & Levy (1967).

A significant development in utilizing the extra degree of freedom was made by Mochrie (1988). He has shown that it is necessary to use the extra degree of freedom in the study of surfaces or interfaces to control the angle of incidence with respect to the surface. His angle calculation scheme for controlling the incident and exit angles for surface scattering has become widely used in surface-scattering studies at synchrotron sources. However, the four-circle geometry still has some limitations for surface-scattering experiments. The accessible solid angle for X-rays is often limited in the surface-scattering chambers. Orienting the sample to a desired direction with respect to resolutions and the polarization of the in-coming X-rays is not, in general, possible. To overcome the geometric limitations of the four-circle diffractometer, a five-circle diffractometer (Gibbs *et al.*, 1990; Vlieg *et al.*, 1987), in which the entire vertically scattering four-circle diffractometer is placed on a rotating table, and a six-circle diffractometer, in which an additional arc (Bloch, 1984) is mounted on the detector arm to provide an extra detector degree of freedom (Lohmeier & Vlieg, 1993; Abernathy, 1995), were developed. A feature of these surface diffractometers, potentially a drawback, is that the detector degrees of freedom are coupled with the sample orientation through the table rotation. Therefore, another type of diffractometer, known as the '2S+2D' diffractometer, was specifically designed for surface-diffraction experiments (Evans-Lutterodt & Tang, 1995). It employs two detector degrees of freedom, both independent of the two sample-orienting circles.

Since three degrees of freedom are needed to determine the orientation of a single-crystal sample, at least three sample-orienting circles are desirable to take full advantage of the two independent detector circles. In fact, the additional extra degree of freedom available with four sample-orienting circles offers the further advantage that one can avoid blind angles, particularly for a diffractometer designed to withstand heavy loading. Therefore, we have developed an angle calculation scheme, described herein, for a diffractometer with four sample-orienting degrees of freedom and two independent detector degrees of freedom (a '4S+2D' six-circle diffractometer). The central idea is based on the capability, with the extra detector degree of freedom, of constraining a reference vector (the surface normal in the surface-diffraction case) to a desired orientation with respect to the laboratory frame of reference. We emphasize, however, that the reference vector is not limited to the surface normal but can be any direction of desired or broken symmetry, either structural, electronic or magnetic in nature.

2. Definition of diffractometer angles and pseudo-angles

The '4S+2D' six-circle diffractometer discussed in this paper is shown schematically in Fig. 1. The notation for the angles is as consistent as possible with the notations previously used with other diffractometers. When the angles μ and ν are set to zero, the diffractometer is essentially a conventional four-circle diffractometer for

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which the θ and 2θ angles are named as η and δ, respectively. The coordinate system for the laboratory frame of reference is chosen to be consistent with that used for the four-circle diffractometer of Busing & Levy (1967) and also with the six-circle surface diffractometer of Lohmeier & Vlieg (1993). It should be noted that one-to-one correspondence with the diffractometer described by Lohmeier & Vlieg (1993) can be made with a set of simple equations, given in the last section (§7) of this paper. Therefore, all the modes described for the six-circle surface diffractometer can be used with the simple transformation and none of those modes of operation will be specifically discussed here. Instead, a general set of equations is developed that allows a user to control the diffractometer with the constraints that most meet the users needs with respect to geometric factors, such as limited accessible solid angles of the sample chamber, the shape, resolution and polarization of the incoming beam, and the orientation of externally applied fields, while still satisfying the Bragg conditions.

We now lay out a simple scattering geometry with angles that relate to the laboratory frame of reference. These angles are the most convenient angles to follow the scattering angles and sample orientations. However, these angles often do not correspond to the diffractometer circles, which are generally based on mechanical considerations. These angles will be used in defining the orientations of vectors such as the scattering vector (**Q**), a reference vector (**n**), or the out-going vector ($\mathbf{k}^f$). Since these angles do not have a one-to-one relation with any diffractometer circles, we will call them pseudo-angles. Some of the pseudo-angles are defined and shown in Fig. 2. The most fundamental pseudo-angle among all the pseudo-angles is θ. In the four-circle case, θ is defined to be half the detector angle, but in the six-circle case it is defined as a pseudo-angle by the scattering equation

$$|\mathbf{Q}| = 2k \sin\theta. \qquad (1)$$

As shown in Fig. 2, the angle θ sets the radius of the sphere that intersects the Ewald sphere. We fix the origin of the laboratory frame of reference at the center of this sphere. We denote the intersecting circle as Ck, which has its plane normal to the incoming beam. It is then evident that the angle of **Q** with respect to the xz plane is θ. We can specify the orientation of **Q** by defining an additional azimuthal angle ϑ (we call it *qaz* for convenience). Similarly, we define two pseudo-angles, α and φ (similarly, *naz*) for the reference vector **n** ($\mathbf{n}' = Q\,\widehat{\mathbf{n}}$ is shown in Fig. 2). When the surface normal is defined to be the reference vector, then α is the incident angle as defined by Mochrie (1988). In addition, two other pseudo-angles, τ and ψ, which are the longitude and azimuth of $\mathbf{n}'$ measured with respect to the scattering vector and scattering plane (a plane formed by the incident beam and the scattering vector), will be used in our calculations. The angle ψ has been previously defined by Mochrie (1988) and originally by Busing & Levy (1967). However τ is a new pseudo-angle, but it is a simple invariance defined by the relation, $\cos\tau = \widehat{\mathbf{Q}} \cdot \widehat{\mathbf{n}}$. Therefore, the only significant new pseudo-angles defined here are the two azimuthal angles φ and ϑ.

3. Coordinate transformation and basic diffraction equation

The format and the angle definitions used in the next two sections were adapted from those of Abernathy (1995), Lohmeier & Vlieg (1993), and Busing & Levy (1967). Some of the algebra is identical or similar to that in the earlier papers but the essential equations are reproduced in order to prepare the readers for the later sections.

Let **h** be the column vector describing a momentum transfer in the reciprocal space with a right-handed coordinate system so that

$$\mathbf{h} = \sum_{i=1}^{3} h_i \mathbf{b}_i, \qquad (2)$$

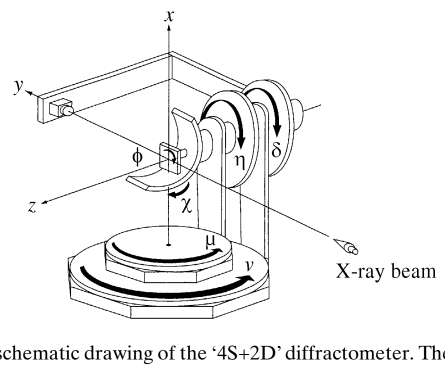

Fig. 1. A schematic drawing of the '4S+2D' diffractometer. The sense of rotations, laboratory frame, and the incoming X-ray directions are shown as arrows.

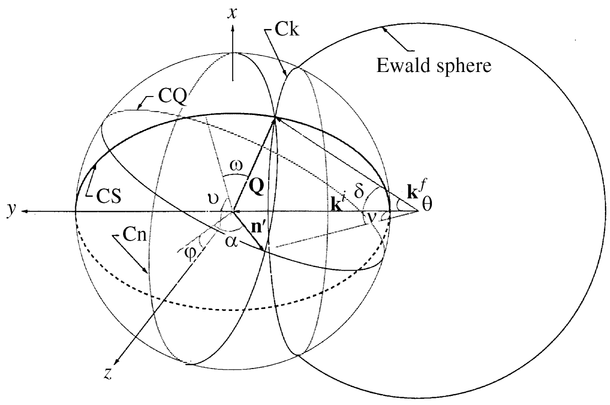

Fig. 2. Definitions of pseudo-angles shown with the scattering vector and Ewald sphere.

where $\mathbf{b}_i$ are the reciprocal-lattice vectors and h_i are generally referred to as h, k and l. Then we can construct a matrix, $\mathbf{B}$, using the Cartesian components of $\mathbf{b}$ in the laboratory frame so that equation (2) can be rewritten as

$$\mathbf{h}_c = \mathbf{B}\mathbf{h}. \tag{3}$$

Note that matrix $\mathbf{B}$ is not necessarily orthonormal as the crystal symmetry is not necessarily cubic. Now let us define the vector $\mathbf{h}_\phi$ which represents the orientation of the reciprocal vector $\mathbf{h}_c$ when the sample is mounted on the diffractometer with every diffractometer circle set to zero. In this case, $\mathbf{h}_\phi$ is simply related to $\mathbf{h}$ by a matrix $\mathbf{U}$, known as the orientation matrix, which corrects the misalignment between the Cartesian axes of the reciprocal crystal space and those of the laboratory frame of reference. The orientation matrix is orthonormal because it simply rotates or reorients the Cartesian coordinate system. Therefore, we can describe $\mathbf{h}_c$ as $\mathbf{h}_j$ after a rotation by the jth sample-orienting circle. Then we can write the relation of $\mathbf{h}$ to $\mathbf{h}_M$ as a simple equation,

$$\mathbf{h} \xrightarrow{\mathbf{B}} \mathbf{h}_c \xrightarrow{\mathbf{U}} \mathbf{h}_\phi \xrightarrow{\mathbf{\Phi}} \mathbf{h}_\chi \xrightarrow{\mathbf{X}} \mathbf{h}_\eta \xrightarrow{\mathbf{H}} \mathbf{h}_\mu \xrightarrow{\mathbf{M}} \mathbf{h}_M, \tag{4}$$

where $\mathbf{\Phi}$, $\mathbf{X}$, $\mathbf{H}$ and $\mathbf{M}$ are the matrices representing the rotation of the corresponding circles. Note that all but $\mathbf{B}$ are orthonormal matrices. The matrices corresponding to the rotation of the circles are explicitly written, using the senses of rotation shown in Fig. 1, as

$$\begin{aligned}
\mathbf{\Phi} &= \begin{pmatrix} \cos\phi & \sin\phi & 0 \\ -\sin\phi & \cos\phi & 0 \\ 0 & 0 & 1 \end{pmatrix},\\
\mathbf{X} &= \begin{pmatrix} \cos\chi & 0 & \sin\chi \\ 0 & 1 & 0 \\ -\sin\chi & 0 & \cos\chi \end{pmatrix},\\
\mathbf{H} &= \begin{pmatrix} \cos\eta & \sin\eta & 0 \\ -\sin\eta & \cos\eta & 0 \\ 0 & 0 & 1 \end{pmatrix},\\
\mathbf{M} &= \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos\mu & -\sin\mu \\ 0 & \sin\mu & \cos\mu \end{pmatrix}.
\end{aligned} \tag{5}$$

Now let us define the detector position in the frame of reference attached to the detector circle δ. When the momentum transfer is zero, the two detector angles are zero and the final outgoing vector is equal to the incoming vector, and is given as

$$\mathbf{k}_0^f = \mathbf{k}^i = \begin{pmatrix} 0 \\ k \\ 0 \end{pmatrix} \tag{6}$$

where the wave number $k = 2\pi/\lambda$. Following a similar scheme used for the six-circle surface diffractometer (Lohmeier & Vlieg, 1993; Abernathy, 1995), this vector is obtained in the laboratory frame of reference for nonzero detector angles by the following two transformations:

$$\mathbf{k}_0^f \xrightarrow{\mathbf{\Delta}} \mathbf{k}_\delta^f \xrightarrow{\mathbf{\Pi}} \mathbf{k}_\nu^f \tag{7}$$

where the explicit forms of $\mathbf{\Delta}$ and $\mathbf{\Pi}$ are

$$\begin{aligned}
\mathbf{\Delta} &= \begin{pmatrix} \cos\delta & \sin\delta & 0 \\ -\sin\delta & \cos\delta & 0 \\ 0 & 0 & 1 \end{pmatrix},\\
\mathbf{\Pi} &= \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos\nu & -\sin\nu \\ 0 & \sin\nu & \cos\nu \end{pmatrix}.
\end{aligned} \tag{8}$$

Note that $\mathbf{H}$, $\mathbf{\Phi}$ and $\mathbf{\Delta}$ are defined by left-handed angles and $\mathbf{M}$, $\mathbf{X}$, $\mathbf{\Phi}$ and $\mathbf{\Pi}$ by right-handed angles. Also note that the x axis is defined along the vertical μ and ν axes and the y axis is defined along the incoming beam direction. Now then

$$\mathbf{k}_\nu^f = k\mathbf{\Pi}\mathbf{\Delta}\begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix} = k\begin{pmatrix} \sin\delta \\ \cos\nu\cos\delta \\ \sin\nu\cos\delta \end{pmatrix}. \tag{9}$$

In order to satisfy the diffraction condition, $\mathbf{h}_M$ should be equal to the desired diffraction vector determined by the detector circles in the laboratory frame of reference. Using the subscript L to emphasize that the vector is the final orientation in the laboratory frame of reference, the diffraction equation becomes

$$\mathbf{h}_M = \mathbf{Q}_L, \tag{10}$$

where

$$\begin{aligned}
\mathbf{h}_M &= \mathbf{MHX\Phi UBh}\\
&= \mathbf{MHX\Phi h}_\phi\\
&= \mathbf{Zh}_\phi,
\end{aligned} \tag{11}$$

described entirely by the sample-orienting circles, and

$$\mathbf{Q}_L = \mathbf{k}_\nu^f - \mathbf{k}_L^i = (\mathbf{\Pi\Delta} - \mathbf{I})\begin{pmatrix} 0 \\ k \\ 0 \end{pmatrix} = k\begin{pmatrix} \sin\delta \\ \cos\delta\cos\nu - 1 \\ \cos\delta\sin\nu \end{pmatrix}, \tag{12}$$

described only by the detector circles. Here $\mathbf{h}_\phi = \mathbf{UBh}$, *i.e.* the lattice vector in the frame of reference attached to the ϕ table of the diffractometer. The total rotational operation made by the sample-orienting circles for the vector transformation, $\mathbf{h}_\phi \rightarrow \mathbf{h}_M$, is

$$
\mathbf{Z} = \mathbf{MH X \Phi}
= \begin{pmatrix}
\begin{matrix}\cos\eta\cos\chi\cos\phi \\ -\sin\eta\sin\phi\end{matrix} & \begin{matrix}\cos\eta\cos\chi\sin\phi \\ +\sin\eta\cos\phi\end{matrix} & \cos\eta\sin\chi \\
\begin{matrix}-\cos\mu\sin\eta \\ \times\cos\chi\cos\phi \\ +\sin\mu\sin\chi\cos\phi \\ -\cos\mu\cos\eta\sin\phi\end{matrix} & \begin{matrix}-\cos\mu\sin\eta \\ \times\cos\chi\sin\phi \\ +\sin\mu\sin\chi\sin\phi \\ +\cos\mu\cos\eta\cos\phi\end{matrix} & \begin{matrix}-\cos\mu\sin\eta\sin\chi \\ -\sin\mu\cos\chi\end{matrix} \\
\begin{matrix}-\sin\mu\sin\eta \\ \times\cos\chi\cos\phi \\ -\cos\mu\sin\chi\cos\phi \\ -\sin\mu\cos\eta\sin\phi\end{matrix} & \begin{matrix}-\sin\mu\sin\eta \\ \times\cos\chi\sin\phi \\ -\cos\mu\sin\chi\sin\phi \\ +\sin\mu\cos\eta\cos\phi\end{matrix} & \begin{matrix}-\sin\mu\sin\eta\sin\chi \\ +\cos\mu\cos\chi\end{matrix}
\end{pmatrix}. \tag{13}
$$

This matrix is used in the later sections for the angle caculations.

4. Motor angles to Miller indices and pseudo-angles

In this section, the calculation of h, k and l from arbitrary detector and sample-orienting angles is given. The same equation can be used to set the orientation matrix when h, k and l are known for particular reflections. From equations (10) and (12), the value of $\mathbf{h}_\phi$ satisfying the diffraction condition is found to be given by

$$
\begin{aligned}
\mathbf{h}_\phi &= \mathbf{Z}^{-1}\mathbf{Q}_L \\
&= k\begin{pmatrix} A\cos\phi\cos\chi + B\sin\phi - C\cos\phi\sin\chi \\ A\sin\phi\cos\chi - B\cos\phi - C\sin\phi\sin\chi \\ A\sin\chi + C\cos\chi \end{pmatrix},
\end{aligned} \tag{14}
$$

where

$$
\begin{aligned}
A &= \sin\eta(\cos\mu - \cos\mu\cos\delta\cos\nu - \sin\mu\cos\delta\sin\nu) \\
&\quad + \cos\eta\sin\delta, \\
B &= \cos\eta(\cos\mu - \cos\mu\cos\delta\cos\nu - \sin\mu\cos\delta\sin\nu) \\
&\quad - \sin\eta\sin\delta, \\
C &= \cos\mu\cos\delta\sin\nu - \sin\mu\cos\delta\cos\nu + \sin\mu.
\end{aligned} \tag{15}
$$

Then, from the definition

$$
\mathbf{h} = (\mathbf{UB})^{-1}\mathbf{h}_\phi, \tag{16}
$$

we can calculate the components h_i (h, k and l) for a given orienation, or when h, k and l are known for two or more reflections, we can determine the orientation matrix.

Now we shall show how to find the pseudo-angles from motor angles. The most fundamental pseudo-angles are θ and ϑ, since the scattering vector $\mathbf{Q}$ can be written either in terms of pseudo-angles or the motor angles. Therefore, $\mathbf{Q}$ alone produces the relation

$$
\widehat{\mathbf{Q}}_L = \frac{\mathbf{Q}_L}{|\mathbf{Q}_L|} = \frac{1}{2\sin\theta}\begin{pmatrix} \sin\delta \\ \cos\delta\cos\nu - 1 \\ \cos\delta\sin\nu \end{pmatrix} \tag{17}
$$

or

$$
\widehat{\mathbf{Q}}_L = \begin{pmatrix} \cos\theta\sin\vartheta \\ -\sin\theta \\ \cos\theta\cos\vartheta \end{pmatrix}, \tag{18}
$$

and from this relation we obtain

$$
\begin{aligned}
\cos 2\theta &= \cos\delta\cos\nu, \\
\tan\vartheta &= \tan\delta/\sin\nu,
\end{aligned} \tag{19}
$$

where $\cos\vartheta$ is zero when ν is zero. The pseudo-angles α and φ can be found from a similar equation for the reference vector,

$$
\widehat{\mathbf{n}}_L = \mathbf{Z}\widehat{\mathbf{n}}_\phi = \begin{pmatrix} \cos\alpha\sin\varphi \\ -\sin\alpha \\ \cos\alpha\cos\varphi \end{pmatrix}. \tag{20}
$$

From this equation we obtain

$$
\sin\alpha = -\widehat{\mathbf{n}}_L \cdot \widehat{\mathbf{y}}_L, \tag{21}
$$

$$
\tan\varphi = \frac{\widehat{\mathbf{n}}_L \cdot \widehat{\mathbf{x}}_L}{\widehat{\mathbf{n}}_L \cdot \widehat{\mathbf{z}}_L}. \tag{22}
$$

Using equations (18) and (20), the angle τ can be determined from the identity $\widehat{\mathbf{n}}_c \cdot \widehat{\mathbf{h}}_c = \widehat{\mathbf{n}}_L \cdot \widehat{\mathbf{h}}_L$ as

$$
\cos\tau = \cos\alpha\cos\theta\cos(\varphi - \vartheta) + \sin\alpha\sin\theta. \tag{23}
$$

Since α is the incident angle when $\mathbf{n}$ is chosen to be the surface normal in surface-scattering experiments, we can conveniently define another pseudo-angle, the exit angle β, from

$$
\sin\beta = \mathbf{k}^f \cdot \mathbf{n}/k^f n.
$$

From the definitions of α and β, we can see that

$$
\begin{aligned}
\sin\alpha + \sin\beta &= [(\mathbf{k}^f - \mathbf{k}^i)/k] \cdot \widehat{\mathbf{n}} \\
&= 2\sin\theta(\widehat{\mathbf{Q}} \cdot \widehat{\mathbf{n}}) \\
&= 2\sin\theta\cos\tau.
\end{aligned} \tag{24}
$$

Therefore,

$$
\sin\beta = 2\sin\theta\cos\tau - \sin\alpha.
$$

We can now imagine a new primed frame of reference where the vector $\mathbf{Q}$ lies along the x' axis and $\mathbf{Q} \times \widehat{\mathbf{y}}$ is along the z' axis in order to define the remaining pseudo-angle ψ. In this frame the z' axis is normal to the scattering plane. We define the zero of the azimuth angle ψ to be when the reference vector $\mathbf{n}$ lies in the scattering plane on the side closer to the $+y'$ axis. Under this condition, the vector $\mathbf{n}'$ points to the intersection of circles CQ and CS as shown in Fig. 2. By writing vectors in the $\mathbf{Q}$ frame of reference as

Table 1. *The five columns of angles used in the various modes of operation discussed in the text (see §5)*

Detector	Reference	Sample	Sample	Sample
δ	$\alpha = \beta$	μ	μ	μ
ν	α	η	η	η
ϑ	β	χ	χ	χ
φ	ψ	ϕ	ϕ	ϕ

$$\widehat{\mathbf{k}}_Q^i = \begin{pmatrix} -\sin\theta \\ \cos\theta \\ 0 \end{pmatrix}, \quad \widehat{\mathbf{k}}_Q^f = \begin{pmatrix} \sin\theta \\ \cos\theta \\ 0 \end{pmatrix},$$
$$\widehat{\mathbf{n}}_Q = \begin{pmatrix} \cos\tau \\ \sin\tau\cos\psi \\ \sin\tau\sin\psi \end{pmatrix}, \qquad (25)$$

the incident and exit angles are determined from the azimuth by

$$\sin\alpha = -\widehat{\mathbf{k}}^i \cdot \widehat{\mathbf{n}} = \cos\tau\sin\theta - \cos\theta\sin\tau\cos\psi \qquad (26)$$

and

$$\sin\beta = \widehat{\mathbf{k}}^f \cdot \widehat{\mathbf{n}} = \cos\tau\sin\theta + \cos\theta\sin\tau\cos\psi, \qquad (27)$$

or conversely, the azimuth ψ can be obtained as

$$\cos\psi = \frac{\cos\tau\sin\theta - \sin\alpha}{\sin\tau\cos\theta} \qquad (28)$$

or

$$\cos\psi = \frac{-\cos\tau\sin\theta + \sin\beta}{\sin\tau\cos\theta}, \qquad (29)$$

as derived from the above equations. This completes the calculation of Miller indices and all the relevant pseudo-angles from only motor angles and a given reference vector.

5. Reciprocal space to angles

Since the diffractometer has six degrees of freedom and determination of a sample orientation in the laboratory frame of reference requires only three angular degrees of freedom, the three remaining angles need to be given in advance. Because of the extra degrees of freedom, there are many modes of diffractometer operation, far more than in the case of a four-circle diffractometer. These modes can conveniently be described by using Table 1, although this is not a unique way of describing the various modes of operation. In Table 1, the first two columns of angles are associated with the detector circles and pseudo-angles and the last three identical columns relate to the sample-orienting circles.

Each mode of operation requires that three columns be chosen out of the five columns of angles shown, and that one angle in each chosen column be given. Of course, one may not choose a particular sample-orienting angle more than once. Some of these modes will be discussed explicitly below.

5.1. *One of the detector angles is given*

The first three angles in the 'detector' column of Table 1, δ, ν and ϑ, are for the detector circles. Given any one of these angles, only one detector degree of freedom remains. Since the angle θ is known from equation (1), both detector angles can be determined by inverting equation (19). Either ν or δ given in advance leads to a quasi-horizontal (δ is frozen) or vertical (ν is frozen) scattering geometry. If the value of the frozen angle is zero, the configuration becomes a true horizontal or vertical scattering mode. As one can see, the pseudo angle ϑ sets the azimuthal angle of the scattering plane. In other words, the scattering plane is horizontal for $\vartheta = 0$ and vertical for $\vartheta = \pi/2$. When one of the detector angles is given, one needs to provide another pseudo-angle and one sample-orienting circle (see §5.3), or two sample-orienting circles (see §5.4) to complete the determination of angular positions for all six circles.

5.2. *One or two of the pseudo-angles for the reference vector are given*

As shown in Fig. 2, the reference vector **n** can point anywhere on the circle CQ, which requires that the pseudo angle ψ be constrained to determine the orientation of the sample crystal uniquely. Alternatively, the pseudo-angles α or φ can be constrained from equation (26) and equation (23). However, one needs to be cautious when the pseudo-angles α or φ are given instead of ψ because their available angular range is limited due to their rotation axes not neccessarilly being aligned with **Q**. These cases belong to the modes where one pseudo-angle and two sample-orienting circles are given, and are discussed in §5.4.

Alternatively, both α and φ can be given in advance, thereby predefining the orientation of the reference vector in the laboratory frame. In this case, $\mathbf{Q}_L$ can be found at the intersections of the circles Ck and Cn. The point of the intersection can be uniquely determined since the pseudo-angle ϑ can be found using the identity equation (23):

$$\cos(\varphi - \vartheta) = \frac{\cos\tau - \sin\alpha\sin\theta}{\cos\alpha\cos\theta}. \qquad (30)$$

Once ϑ is known, the detector angles are determined from equation (19). Alternatively α and ϑ can simply be given instead. In either case, with the two pseudo-angles, one sample orienting circle needs to be given in advance to determine the sample orientation. This case is discussed in §5.3.

5.3. *When one sample-orienting angle and two other angles are given*

Two angles from the first two columns of Table 1 will uniquely set the detctor position for a given scattering vector and orientation of the reference vector. There is only one sample orientation satisfying the orientations of both the scattering and reference vectors (as long as they are not collinear). So three sample degrees of freedom must be used to orient the sample. Therefore the position of one sample-orienting circle must be given in advance.

The vectors $\widehat{\mathbf{n}}$ and $\widehat{\mathbf{Q}}$ are fixed in the laboratory frame as defined in equations (18) and (20). An orthonormal matrix ($\mathbf{N}_L$) based on these two vectors can be constructed by the three vectors $\widehat{\mathbf{Q}}$, $\widehat{\mathbf{Q}}\times\widehat{\mathbf{n}}\times\mathbf{Q}$ and $\widehat{\mathbf{Q}}\times\widehat{\mathbf{n}}$ [known as Gram–Schmidt orthonormalization; also used by Busing & Levy (1967)] in the laboratory frame of reference as

$$\mathbf{N}_L=\begin{pmatrix} \widehat{\mathbf{Q}}\Big|_1 & \widehat{\mathbf{Q}}\times\widehat{\mathbf{n}}\times\widehat{\mathbf{Q}}\Big|_1 & \widehat{\mathbf{Q}}\times\widehat{\mathbf{n}}\Big|_1 \\ \widehat{\mathbf{Q}}\Big|_2 & \widehat{\mathbf{Q}}\times\widehat{\mathbf{n}}\times\widehat{\mathbf{Q}}\Big|_2 & \widehat{\mathbf{Q}}\times\widehat{\mathbf{n}}\Big|_2 \\ \widehat{\mathbf{Q}}\Big|_3 & \widehat{\mathbf{Q}}\times\widehat{\mathbf{n}}\times\widehat{\mathbf{Q}}\Big|_3 & \widehat{\mathbf{Q}}\times\widehat{\mathbf{n}}\Big|_3 \end{pmatrix}, \tag{31}$$

where $\mathbf{a}|_1$ indicates the normalized x component of vector $\mathbf{a}$, *i.e.* $a_x/|a|$. This matrix is related to another orthonormal matrix ($\mathbf{N}_\phi$) formed by the corresponding two vectors $\mathbf{h}_\phi$ and $\mathbf{n}_\phi$ in the ϕ frame through the matrix $\mathbf{Z}$ as

$$\mathbf{N}_L=\mathbf{Z}\mathbf{N}_\phi \tag{32}$$

when $\mathbf{N}_\phi$ is constructed from $\mathbf{h}_\phi$ and $\mathbf{n}_\phi$ (components are known) in an identical manner. Since $\mathbf{Z}=\mathbf{N}_L\mathbf{N}_\phi^{-1}$ from equation (32), the sample-orienting circles are determined by comparing the matrix components in the following relations:

$$\begin{aligned} \mathbf{HX\Phi} &= \mathbf{M}^{-1}\mathbf{N}_L\mathbf{N}_\phi^{-1} \quad \text{for } \mu \text{ fixed,} \\ \mathbf{MHX} &= \mathbf{N}_L\mathbf{N}_\phi^{-1}\mathbf{\Phi}^{-1} \quad \text{for } \phi \text{ fixed,} \\ \mathbf{HX\Phi} &= \mathbf{M}^{-1}\mathbf{N}_L\mathbf{N}_\phi^{-1} \quad \text{for } \eta \text{ fixed.} \end{aligned} \tag{33}$$

For μ fixed ($\mu=\mu_0$ or $\nu/2$), $\mathbf{V}=\mathbf{M}^{-1}\mathbf{N}_L\mathbf{N}_\phi^{-1}$ is known, and its components are to be compared to

$$\mathbf{HX\Phi}=\begin{pmatrix} \cdots & \cdots & \cos\eta\sin\chi \\ \cdots & \cdots & -\sin\eta\sin\chi \\ -\sin\chi\cos\phi & -\sin\chi\sin\phi & \cos\chi \end{pmatrix}. \tag{34}$$

Only the matrix components explicitly shown above are needed to determine ϕ, χ and η [in the same manner as Busing & Levy (1967)]:

$$\begin{aligned} \phi &= \arctan(V_{32}, V_{31}), \\ \eta &= \arctan(-V_{23}, V_{13}), \\ \chi &= \arctan[(V_{31}^2+V_{32}^2)^{1/2}, V_{33}]; \quad -\pi/2<\chi<\pi/2, \\ &= \arccos(V_{33}); \quad 0<\chi<\pi. \end{aligned} \tag{35}$$

Likewise, for ϕ fixed, $\mathbf{V}=\mathbf{N}_L\mathbf{N}_\phi^{-1}\mathbf{\Phi}^{-1}$ is known and compared to

$$\mathbf{MHX}=\begin{pmatrix} \cos\eta\cos\chi & \sin\eta & \cos\eta\sin\chi \\ \cdots & \cos\mu\cos\eta & \cdots \\ \cdots & \sin\mu\cos\eta & \cdots \end{pmatrix} \tag{36}$$

with μ, χ and η similarly determined as

$$\begin{aligned} \eta &= \arctan[V_{12}, (V_{22}^2+V_{32}^2)^{1/2}], \\ \mu &= \arctan(V_{32}, V_{22}), \\ \chi &= \arctan(V_{13}, V_{11}); \quad -\pi/2<\chi<\pi/2, \\ &= \arccos(V_{11}/\cos\eta); \quad 0<\chi<\pi. \end{aligned} \tag{37}$$

For η fixed ($\eta=\eta_0$ or $\eta=\delta/2$) or χ fixed, $\mathbf{V}=\mathbf{N}_L\mathbf{N}_\phi^{-1}$ is known and needs to be compared to

$$\mathbf{Z}=\mathbf{MHX\Phi}=\begin{pmatrix} \begin{matrix}\cos\eta\cos\chi\cos\phi \\ -\sin\eta\sin\phi\end{matrix} & \begin{matrix}\cos\eta\cos\chi\sin\phi \\ +\sin\eta\cos\phi\end{matrix} & \cos\eta\sin\chi \\ \cdots & \cdots & \begin{matrix}-\cos\mu\sin\eta\sin\chi \\ -\sin\mu\cos\chi\end{matrix} \\ \cdots & \cdots & \begin{matrix}-\sin\mu\sin\eta\sin\chi \\ +\cos\mu\cos\chi\end{matrix} \end{pmatrix}. \tag{38}$$

From the comparison of V_{13}, χ and η are simply determined as

$$\chi=\arcsin(V_{13}/\cos\eta); \quad -\pi/2<\chi<\pi/2 \tag{39}$$

and

$$\eta=\arccos(V_{13}/\sin\chi); \quad 0<\chi<\pi. \tag{40}$$

The other two angles, μ and ϕ, are similarly determined by examining the (V_{33}, V_{23}) and (V_{11}, V_{12}) pairs:

$$\mu=\arctan\left(\frac{V_{33}\sin\eta\sin\chi+V_{23}\cos\chi}{-V_{33}\cos\chi+V_{23}\sin\eta\sin\chi}\right) \tag{41}$$

and

$$\phi=\arctan\left(\frac{V_{12}\cos\eta\cos\chi-V_{11}\sin\eta}{V_{12}\sin\eta+V_{11}\cos\eta\cos\chi}\right). \tag{42}$$

5.4. *When two sample-orienting angles and one detector or one pseudo-angle are given*

For the case in which two sample-orienting angles and one of the angles in the first two columns of Table 1 are given, it is desirable that the frame attached to the scattering and reference vectors is specified by successive rotations of the laboratory frame about the x axis by ψ, then about the z axis by $-\theta$, and finally about the y axis by $\pi/2 - \vartheta$. Then the orthonormal matrix is $\mathbf{N}_L = \boldsymbol{F}\boldsymbol{\Theta}\boldsymbol{\Psi}$, and we rewrite equation (32) as

$$\boldsymbol{F}\boldsymbol{\Theta}\boldsymbol{\Psi} = \mathbf{MHX}\boldsymbol{\Phi}\mathbf{N}_\phi \tag{43}$$

where

$$\boldsymbol{\Theta} = \begin{pmatrix} \cos\theta & \sin\theta & 0 \\ -\sin\theta & \cos\theta & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad \boldsymbol{\Psi} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos\psi & -\sin\psi \\ 0 & \sin\psi & \cos\psi \end{pmatrix} \tag{44}$$

and

$$\boldsymbol{F} = \begin{pmatrix} \cos\xi & 0 & \sin\xi \\ 0 & 1 & 0 \\ -\sin\xi & 0 & \cos\xi \end{pmatrix}, \tag{45}$$

with $\xi = \vartheta - \pi/2$.

For the case in which χ, ϕ and ψ are given, it is convenient to rewrite equation (43) as

$$\mathbf{H}^{-1}\mathbf{M}^{-1}\boldsymbol{F} = \mathbf{X}\boldsymbol{\Phi}\mathbf{N}_\phi\boldsymbol{\Psi}^{-1}\boldsymbol{\Theta}^{-1}. \tag{46}$$

The variables on the right-hand side are all known and we call the right-hand side $\mathbf{V}$. Now let us express the left-hand side in terms of the unknown angles η, ϑ and μ as

$$\mathbf{H}^{-1}\mathbf{M}^{-1}\boldsymbol{F} = \begin{pmatrix} \cdots & -\sin\eta\cos\mu & \cdots \\ \cdots & \cos\eta\cos\mu & \cdots \\ -\cos\mu\sin\xi & -\sin\mu & \cos\mu\cos\xi \end{pmatrix}. \tag{47}$$

Its components should then be equal to the components of $\mathbf{V} = \mathbf{X}\boldsymbol{\Phi}\mathbf{N}_\phi\boldsymbol{\Psi}^{-1}\boldsymbol{\Theta}^{-1}$. By comparing, we obtain

$$\begin{aligned} \xi &= \arctan(-V_{31}, V_{33}), \\ \eta &= \arctan(-V_{12}, V_{22}), \\ \mu &= \arctan[-V_{32}, (V_{33}^2 + V_{31}^2)^{1/2}]; \quad -\pi/2<\mu<\pi/2, \\ &= \arctan[-V_{32}, (V_{12}^2 + V_{22}^2)^{1/2}]; \quad -\pi/2<\mu<\pi/2, \\ &= \arcsin(-V_{32}); \quad -\pi/2<\mu<\pi/2. \end{aligned} \tag{48}$$

When ψ, μ and η are given, the following form is most useful:

$$\boldsymbol{\Phi}^{-1}\mathbf{X}^{-1}\mathbf{H}^{-1}\mathbf{M}^{-1}\boldsymbol{F} = \mathbf{N}_\phi\boldsymbol{\Psi}^{-1}\boldsymbol{\Theta}^{-1}. \tag{49}$$

We again define the right-hand side as $\mathbf{V}$. Then the left-hand side becomes

$$\boldsymbol{\Phi}^{-1}\mathbf{X}^{-1}\mathbf{H}^{-1}\mathbf{M}^{-1}\boldsymbol{F} = \begin{pmatrix} \cdots & \begin{matrix} -\cos\mu\cos\phi \\ \times\cos\chi\sin\eta \\ -\cos\mu\sin\phi\cos\eta \\ +\cos\phi\sin\chi\sin\mu \end{matrix} & \cdots \\ \cdots & \begin{matrix} -\cos\mu\sin\phi \\ \times\cos\chi\sin\eta \\ +\cos\mu\cos\phi\cos\eta \\ +\sin\phi\sin\chi\sin\mu \end{matrix} & \cdots \\ \begin{matrix} \sin\chi\cos\eta\cos\xi \\ +\sin\xi\sin\chi \\ \times\sin\eta\sin\mu \\ -\sin\xi\cos\chi\cos\mu \end{matrix} & \begin{matrix} -\sin\chi\sin\eta\cos\mu \\ -\cos\chi\sin\mu \end{matrix} & \begin{matrix} \sin\chi\cos\eta\sin\xi \\ -\cos\xi\sin\chi \\ \times\sin\eta\sin\mu \\ +\cos\xi\cos\chi\cos\mu \end{matrix} \end{pmatrix}. \tag{50}$$

These components should match those of $\mathbf{V}$. By comparing V_{32} to the corresponding component of this matrix, we have

$$-V_{32} = \sin\chi\sin\eta\cos\mu + \cos\chi\sin\mu. \tag{51}$$

Since we know the values of η and μ,

$$\chi = \arcsin\left[\frac{-V_{32}}{(\sin^2\eta\cos^2\mu + \sin^2\mu)^{1/2}}\right] - \arctan\left(\frac{\sin\mu}{\sin\eta\cos\mu}\right). \tag{52}$$

An examination of the matrix components V_{31} and V_{33} yields

$$\frac{V_{31}\cos\xi + V_{33}\sin\xi}{V_{31}\sin\xi - V_{33}\cos\xi} = \frac{\sin\chi\cos\eta}{\sin\chi\sin\eta\sin\mu - \cos\chi\cos\mu} \tag{53}$$

and from this equation we obtain ξ using the value of χ obtained above,

$$\xi = \arctan\left[\frac{V_{33}\sin\chi\cos\eta + V_{31}(\sin\chi\sin\eta\sin\mu - \cos\chi\cos\mu)}{V_{31}\sin\chi\cos\eta - V_{33}(\sin\chi\sin\eta\sin\mu - \cos\chi\cos\mu)}\right]. \tag{54}$$

We can similarly solve for ϕ from the matrix components V_{12} and V_{22} also using the value of χ.

$$\phi = \arctan\left[\frac{V_{22}(\sin\chi\sin\mu - \cos\mu\cos\chi\sin\eta) - V_{12}\cos\mu\cos\eta}{V_{12}(\sin\chi\sin\mu - \cos\mu\cos\chi\sin\eta) + V_{22}\cos\mu\cos\eta}\right]. \tag{55}$$

When ϑ, η and μ are given, the equation becomes

$$\mathbf{H}^{-1}\mathbf{M}^{-1}\boldsymbol{F}\boldsymbol{\Theta} = \mathbf{X}\boldsymbol{\Phi}\mathbf{N}_\phi\boldsymbol{\Psi}^{-1}, \tag{56}$$

where all the components of the left-hand side are known and we again call it $\mathbf{V}$. Then the right-hand side is

$$\mathbf{X\Phi N}_\phi\mathbf{\Psi}^{-1} = \begin{pmatrix} (\cos\chi\cos\phi)N_{11} + (\cos\chi\sin\phi)N_{21} + (\sin\chi)N_{31} & \cdots & \cdots \\ -(\sin\phi)N_{11} + (\cos\phi)N_{21} & \begin{matrix}-(\cos\psi\sin\phi)N_{12}\\ +(\cos\psi\cos\phi)N_{22}\\ +(\sin\psi\sin\phi)N_{13}\\ -(\sin\psi\cos\phi)N_{23}\end{matrix} & \begin{matrix}-(\sin\psi\sin\phi)N_{12}\\ +(\sin\psi\cos\phi)N_{22}\\ -(\cos\psi\sin\phi)N_{13}\\ +(\cos\psi\cos\phi)N_{23}\end{matrix} \\ -(\sin\chi\cos\phi)N_{11} - (\sin\chi\sin\phi)N_{21} + (\cos\chi)N_{31} & \cdots & \cdots \end{pmatrix}. \tag{57}$$

By comparing the V_{21} component we can see that

$$V_{21} = -N_{11}\sin\phi + N_{21}\cos\phi. \tag{58}$$

Therefore we obtain

$$\phi = \arcsin\left[-\frac{V_{21}}{(N_{11}^2 + N_{21}^2)^{1/2}}\right] + \arctan\left(\frac{N_{21}}{N_{11}}\right). \tag{59}$$

As performed above, an examination of the displayed components yields

$$\chi = \arctan\left[\frac{N_{31}V_{11} - V_{31}(N_{11}\cos\phi + N_{21}\sin\phi)}{N_{31}V_{31} + V_{11}(N_{11}\cos\phi + N_{21}\sin\phi)}\right] \tag{60}$$

and

$$\psi = \arctan\left[\frac{N_{12}V_{23}\tan\phi - N_{13}V_{22}\tan\phi + N_{23}V_{22} - N_{22}V_{23}}{V_{23}(N_{13}\tan\phi - N_{23}) + V_{22}(N_{12}\tan\phi + N_{22})}\right]. \tag{61}$$

For given ϕ, χ and $\vartheta = \xi + \pi/2$, the equation becomes

$$\mathbf{H}^{-1}\mathbf{M}^{-1}\boldsymbol{F}\mathbf{\Theta\Psi} = \mathbf{X\Phi N}_\phi. \tag{62}$$

From the left-hand side we obtain

$$\mathbf{H}^{-1}\mathbf{M}^{-1}\boldsymbol{F}\mathbf{\Theta\Psi} = \begin{pmatrix} \begin{matrix}\cos\theta\cos\eta\cos\xi\\ +\cos\theta\sin\eta\\ \times\sin\mu\sin\xi\\ +\sin\eta\cos\mu\sin\theta\end{matrix} & \cdots & \cdots \\ \begin{matrix}\cos\theta\sin\eta\cos\xi\\ -\cos\theta\cos\eta\\ \times\sin\mu\sin\xi\\ -\cos\eta\cos\mu\sin\theta\end{matrix} & \cdots & \cdots \\ \begin{matrix}-\cos\mu\sin\xi\cos\theta\\ +\sin\mu\sin\theta\end{matrix} & \begin{matrix}-\cos\psi\cos\mu\\ \times\sin\xi\sin\theta\\ -\cos\psi\sin\mu\cos\theta\\ +\cos\mu\cos\xi\sin\psi\end{matrix} & \begin{matrix}\sin\psi\cos\mu\\ \times\sin\xi\sin\theta\\ +\sin\psi\sin\mu\cos\theta\\ +\cos\mu\cos\xi\cos\psi\end{matrix} \end{pmatrix}. \tag{63}$$

By defining the right-hand side as V, we can similarly solve for η, μ and ψ as before. By comparing the V_{31} component, we find

$$\mu = \arcsin\left[\frac{V_{31}}{(\sin^2\xi\cos^2\theta + \sin^2\theta)^{1/2}}\right] + \arctan\left(\frac{\sin\xi\cos\theta}{\sin\theta}\right). \tag{64}$$

From the ratio of V_{11} and V_{21}, we can obtain the value of η,

$$\eta = \arctan\left[\frac{V_{21}\cos\theta\cos\xi + V_{11}(\cos\theta\sin\mu\sin\xi + \cos\mu\sin\theta)}{V_{11}\cos\theta\cos\xi - V_{21}(\cos\theta\sin\mu\sin\xi + \sin\eta\cos\mu\sin\theta)}\right]. \tag{65}$$

Similarly, we obtain ψ from the ratio of V_{32} and V_{33},

$$\psi = \arctan\left[\frac{-V_{33}(\cos\mu\sin\xi\sin\theta + \sin\mu\cos\theta) - V_{32}\cos\mu\cos\xi}{V_{32}(\cos\mu\sin\xi\sin\theta + \sin\mu\cos\theta) - V_{33}\cos\mu\cos\xi}\right]. \tag{66}$$

Other conditions such as fixing the values of ϕ and μ, χ and μ, ϕ and η, or χ and μ along with a pseudo-angle can be similarly obtained.

5.5. *When three sample-orienting angles are given*

Now let us consider the case when three sample-orienting angles are given. Although we see no advantage in using such a mode under normal scattering conditions, one can imagine, nevertheless, a situation where three sample-orienting circles have only limited angular ranges, for example, because of geometric constraint due to a sample chamber. In these modes, the reference vector is not necessary to obtain the solution. There are two ways to solve these cases: solving without the reference vector or solving with a conveniently chosen reference vector as discussed in §6. Since the latter case can be treated in the same way as the previous cases in which two sample-orienting circles are given, we will outline the former case, *i.e.* without using the reference vector.

Here, we use the vector equation

$$\widehat{\mathbf{Q}}_L = \mathbf{MHX\Phi}\widehat{\mathbf{h}}_\phi. \tag{67}$$

From equations (10) and (18) we obtain

$$\begin{pmatrix} \cos\theta\sin\vartheta \\ -\sin\theta \\ \cos\theta\cos\vartheta \end{pmatrix} = \begin{pmatrix} (\cos\eta\cos\chi\cos\phi - \sin\eta\sin\phi)\widehat{h}_{\phi,1} \\ +(\cos\eta\cos\chi\sin\phi + \sin\eta\cos\phi)\widehat{h}_{\phi,2} \\ +(\cos\eta\sin\chi)\widehat{h}_{\phi,3} \\ [(-\cos\mu\sin\eta\cos\chi + \sin\mu\sin\chi)\cos\phi - \cos\mu\cos\eta\sin\phi]\widehat{h}_{\phi,1} \\ +[(-\cos\mu\sin\eta\cos\chi + \sin\mu\sin\chi)\sin\phi + \cos\mu\cos\eta\cos\phi]\widehat{h}_{\phi,2} \\ +(-\cos\mu\sin\eta\sin\chi - \sin\mu\cos\chi)\widehat{h}_{\phi,3} \\ [(-\sin\mu\sin\eta\cos\chi - \cos\mu\sin\chi)\cos\phi - \sin\mu\cos\eta\sin\phi]\widehat{h}_{\phi,1} \\ +[(-\sin\mu\sin\eta\cos\chi - \cos\mu\sin\chi)\sin\phi + \sin\mu\cos\eta\cos\phi]\widehat{h}_{\phi,2} \\ +(-\sin\mu\sin\eta\sin\chi + \cos\mu\cos\chi)\widehat{h}_{\phi,3} \end{pmatrix}. \tag{68}$$

We then rearrange the y component of this equation to the following form:

$$a\sin\varsigma + b\cos\varsigma = c, \tag{69}$$

where ς denotes the unknown angle (one of ϕ, χ, η or μ). The unknown angle can then be found as

$$\varsigma = \arcsin[c/(a^2+b^2)^{1/2}] - \arctan(b,a). \tag{70}$$

Using this angle, we can additionally find

$$\vartheta = \arctan\left(\frac{\begin{matrix}(\cos\eta\cos\chi\cos\phi - \sin\eta\sin\phi)\widehat{h}_{\phi,1} \\ +(\cos\eta\cos\chi\sin\phi + \sin\eta\cos\phi)\widehat{h}_{\phi,2} \\ +(\cos\eta\sin\chi)\widehat{h}_{\phi,3}\end{matrix}}{\begin{matrix}[(-\sin\mu\sin\eta\cos\chi - \cos\mu\sin\chi) \\ \times\cos\phi - \sin\mu\cos\eta\sin\phi]\widehat{h}_{\phi,1} \\ +[(-\sin\mu\sin\eta\cos\chi - \cos\mu\sin\chi) \\ \times\sin\phi + \sin\mu\cos\eta\cos\phi]\widehat{h}_{\phi,2} \\ +(-\sin\mu\sin\eta\sin\chi + \cos\mu\cos\chi)\widehat{h}_{\phi,3}\end{matrix}}\right) \tag{71}$$

with which one can subsequently set the angles for the detector circles according to equation (19).

6. Relation to other diffractometer operation modes

The modes of operation discussed above include most of the known diffractometer settings and modes of operation. Since it may not be apparent how the modes described here correspond to some of the previously known modes of operation, we will discuss the relation of the previously popular modes of diffractometer operation to the modes discussed above.

The most popular mode for the four-circle diffractometer is the so-called $\omega = 0$ mode or bisecting mode. In this mode, the scattering vector is parallel to the plane of the χ circle. For either horizontal or vertical scattering condition, one can reproduce the bisecting mode of the four-circle case simply by setting $\vartheta = 0$ or $\vartheta = 90°$ and by setting $\mu = \nu/2$ or $\eta = \delta/2$, respectively. Unlike the four-circle case, there is an extra degree of freedom available for orienting the sample, such as choosing a desirable ψ, α or ϑ. Therefore, we define ω as the angle of the scattering vector ($\mathbf{Q}$) with respect to the plane of the χ circle. Since the axis of the χ circle is on $\widehat{\mathbf{y}}$ when $\eta = \mu = 0$, we obtain the relation $\widehat{\mathbf{Q}}\cdot(\mathbf{M}\mathbf{H}\widehat{\mathbf{y}}) = \cos(\omega + \pi/2)$ and from this we obtain

$$(\sin\eta\sin\vartheta + \sin\mu\cos\eta\cos\vartheta)\cos\theta - (\cos\mu\cos\eta)\sin\theta = \sin\omega. \tag{72}$$

When the axis of the χ circle is within the scattering plane, as expected for a four-circle diffractometer, we obtain an additional equation,

$$\sin\mu\cos\eta\sin\vartheta = \sin\eta\cos\vartheta \tag{73}$$

from the condition $\widehat{\mathbf{y}}\cdot(\widehat{\mathbf{Q}}\times\mathbf{M}\mathbf{H}\widehat{\mathbf{y}}) = 0$. We can now obtain a set of equations for the angles η and μ from these two equations:

$$\sin\eta = \sin(\theta+\omega)\sin\vartheta \tag{74}$$

and

$$\tan\mu = \tan(\theta+\omega)\cos\vartheta. \tag{75}$$

Another popular mode of operation of the four-circle diffractometer is the zone mode. In this mode, a zone (a plane including the origin of reciprocal space) is specified by two reciprocal-lattice vectors and the specified zone is moved to the scattering plane. In this condition, one can easily navigate within the plane of reciprocal space without changing the orientation of instrumental resolution, the polarization of incoming X-rays or neutrons, or the direction of the external field applied to the sample with respect to the zone. Let us call the two input reciprocal vectors $\mathbf{p}_\phi$ and $\mathbf{r}_\phi$ and define the reference vector $\mathbf{n}_\phi = \mathbf{p}_\phi \times \mathbf{r}_\phi$. The zone mode can be achieved by setting $\varphi = \vartheta \pm \pi/2$ and $\psi = \pi/2$ for a given value of ϑ. This condition constrains $\mathbf{n}_L$ to the xz plane and normal to the scattering plane. There are several ways to satisfy this condition but we will describe a way that is most similar to the zone mode of the four-circle diffractometer. First, we find ϕ and χ that satisfy the conditions $\vartheta = 0$ and $\mathbf{n}_L = \mathbf{Z}\widehat{\mathbf{n}}_\phi \| \widehat{\mathbf{x}}$, as if the diffractometer is a horizontal four-circle diffractometer. From this condition we obtain the known solution:

$$\tan\phi_z = n_2/n_1 \tag{76}$$

and

$$\tan\chi_z = n_3/(n_1^2 + n_2^2)^{1/2}. \tag{77}$$

We can use ϑ, χ_z and ϕ_z to determine the remaining angles, μ and η. For either the horizontal or vertical scattering condition, one can reproduce the zone modes of the four-circle case by setting $\vartheta = 0$ and $\eta = 0$, or $\vartheta = 90$ and $\mu = 0°$. The accessible reciprocal vectors in the zone modes should, of course, be limited to $\mathbf{h}_\phi = a\mathbf{p}_\phi + b\mathbf{r}_\phi$ since reciprocal vectors that are not in the zone can also be reached in this manner due to the extra degree of freedom.

The above two modes are related just as in the four-circle case. Once χ and ϕ are frozen, the vectors in the zone are reached by changing ω only, just as in the four-circle case. The change of ω is ensured by setting ϑ and finding the corresponding η and μ. The η and μ then satisfy the relation given in equation (73).

There are several modes of operation for which the definition of the reference vector $\mathbf{n}$ is not needed. Examples are the modes where ν, δ or ϑ and two of the sample-orienting circles are given. Nevertheless, the pseudo-angles for a specified reference vector can be calculated as long as the scattering vector is not parallel to the reference vector. In order to avoid accidentally setting the reference vector parallel to the scattering vector using an arbitray reference vector, we again use the Gram–Schmidt orthonormalization procedure (see Busing & Levy, 1967) to find a suitable reference vector. The following matrix can be formed solely by the scattering vector $\widehat{\mathbf{q}}$ either in the laboratory frame or in the ϕ frame:

$$\begin{pmatrix} \widehat{q}_1 & 0 & 0 \\ \widehat{q}_2 & 0 & 0 \\ \widehat{q}_3 & 0 & 0 \end{pmatrix} \longrightarrow \begin{pmatrix} \widehat{q}_1 & (\widehat{q}_2^2+\widehat{q}_3^2)^{1/2} & 0 \\ \widehat{q}_2 & -\frac{\hat{q}_1\hat{q}_2}{(\hat{q}_2^2+\hat{q}_3^2)^{1/2}} & \frac{\hat{q}_3}{(\hat{q}_2^2+\hat{q}_3^2)^{1/2}} \\ \widehat{q}_3 & -\frac{\hat{q}_1\hat{q}_3}{(\hat{q}_2^2+\hat{q}_3^2)^{1/2}} & -\frac{\hat{q}_2}{(\hat{q}_2^2+\hat{q}_3^2)^{1/2}} \end{pmatrix}. \tag{78}$$

We can set the second (or third) column of this matrix to be the reference vector $\mathbf{n}$.

7. Relation to the six-circle surface diffractometer

In this section, we derive the relation between the motors of the six-circle surface diffractometer and our '4S+2D' type of six-circle diffractometer. A part of the algebra has essentially been reported by Vlieg (1998) but is reproduced here with appropriate notations for clarity. Note that the two diffractometers differ only by the detector circles. That is, the circles γ and δ' for the surface diffractometer sit on the μ table, thereby coupling the γ and δ' rotations to the μ rotation. The final wavevector $\widehat{\mathbf{k}}_f$ in the surface diffractometer is given by

$$\begin{aligned} \widehat{\mathbf{k}}_f &= \begin{pmatrix} \sin\delta'\cos\gamma \\ \cos\delta'\cos\mu\cos\gamma - \sin\mu\sin\gamma \\ \cos\delta'\sin\mu\cos\gamma + \cos\mu\sin\gamma \end{pmatrix} \\ &\equiv \begin{pmatrix} \sin\delta \\ \cos\delta\cos\nu \\ \cos\delta\sin\nu \end{pmatrix}. \end{aligned} \tag{79}$$

From this identity we obtain the relations

$$\sin\gamma = \cos\delta\sin(\nu-\mu), \tag{80}$$

$$\tan\delta' = \tan\delta/\cos(\nu-\mu) \tag{81}$$

and

$$\sin\delta' = \sin\delta/\cos\gamma, \tag{82}$$

to convert from the '4S+2D'-type six-circle diffractometer to the six-circle surface diffractometer, or

$$\sin\delta = \sin\delta'\cos\gamma \tag{83}$$

and

$$\tan\nu = \frac{\cos\delta'\cos\gamma\sin\mu + \cos\mu\sin\gamma}{\cos\delta'\cos\gamma\cos\mu - \sin\mu\sin\gamma} \tag{84}$$

to convert from the six-circle surface diffractometer to the '4S+2D'-type six-circle diffractometer.

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